VOL. 258 DECEMBER 1960 No. 10

American Journal of Science

Established in 1818 by Benjamin Silliman

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Published 10 times a year. Ten dollars a year. One dollar twenty-five cents a copy.
Foreign, ten dollars sixty cents. Canada, ten dollars thirty-five cents.

AMERICAN JOURNAL OF SCIENCE

Established in 1818 by Benjamin Silliman

THE FIRST SCIENTIFIC JOURNAL IN THE UNITED STATES

DEVOTED TO THE GEOLOGICAL SCIENCES AND TO RELATED FIELDS

Editors: JOHN RODGERS—JOSEPH T. GREGORY
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Entered as second-class matter at the Post Office at New Haven, Conn., under the Act of March 3, 1879. Published monthly, except July and September, at Sterling Tower, Yale University, New Haven, Conn.

Subscription rate $10 per year ($1.25 a number). Student rate $5 per year (application blanks available on request). Postage prepaid to the United States; 35 cents per year to Canada; 60 cents per year elsewhere.

The JOURNAL completed its *first* series of 50 volumes in 1845, its *second* series of 50 volumes in 1870, its *third* series of 50 volumes in 1895, its *fourth* series of 50 volumes in 1920, its *fifth* series of 36 volumes in 1938. Since 1938 the numbering by series has been discontinued, and volumes are annual and bear the whole numbers as of 1818, that for 1939 being volume 237.

Back numbers and volumes, also cumulative indices of all series, may with few exceptions be obtained from the office of the AMERICAN JOURNAL OF SCIENCE, Sterling Tower, New Haven, Conn. Positive microfilm copies of each complete volume may be purchased by subscribers to the paper edition from *University Microfilms*, 313 N. First St., Ann Arbor, Michigan. Regular subscribers may purchase each past year in microcard form, from *The Microcard Foundation*, Wesleyan University, Middletown, Connecticut.

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Suggestions to Authors of the Reports of the United States Geological Survey, 5th ed. Washington, D. C., 1958 (Government Printing Office, $1.75).

[American Journal of Science, Vol. 258, December 1960, P. 689-704]

American Journal of Science

DECEMBER 1960

LAWSONITE AND PUMPELLYITE IN GLAUCOPHANE SCHIST, NORTH BERKELEY HILLS, CALIFORNIA

GREGORY A. DAVIS

WITH NOTES ON THE X-RAY CRYSTALLOGRAPHY OF LAWSONITE

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ABSTRACT. A previously undescribed lawsonite-pumpellyite locality in the North Berkeley Hills is characterized by exceptional development of lawsonite as veins and as lithologic layers in glaucophane schist. The glaucophane schist outcrop, a tectonic block in serpentinite, is believed to have been derived at least in part by retrogressive metamorphism of eclogite. Lawsonite-rich layers in the schists and the vein minerals lawsonite and pumpellyite are regarded as products of metamorphic differentiation under conditions of glaucophanitic metamorphism.

Optical and morphological data for lawsonite and pumpellyite are presented together with a new lawsonite chemical analysis and a semi-quantitative spectroscopic analysis of pumpellyite. A description is given of lawsonite-pumpellyite intergrowths and of pumpellyite twinning.

The cell dimensions of lawsonite from three California localities and from Cuba have been determined by means of calibrated Weissenberg patterns. All values obtained lie within the limits 8.788 ± 0.008 Å for a_o, 13.129 ± 0.010 for b_o and 5.846 ± 0.006 for c_o. A powder diffraction pattern giving calculated and observed d values to 1.46 Å has been tabulated. The thermal transformation of lawsonite to metastable hexagonal $CaAl_2Si_2O_8$ has been studied by means of single crystal diffraction. The change can be detected after prolonged heating at temperatures as low as 410°C. In the neighborhood of 700°C it is complete in an hour or less. At intermediate temperatures partly changed lawsonite may persist for long periods. Single crystal patterns are more sensitive than powder patterns for detecting the beginning and end of the transformation. Lawsonite that had been heated under a bonfire was found to be partly transformed.

INTRODUCTION

Glaucophane schists from the North Berkeley Hills and associated occurrences of lawsonite and pumpellyite have been described by Brothers (1954). One lawsonite-pumpellyite-glaucophane schist locality not observed by Brothers has been found on the private estate of Mrs. Anson Blake and the late Mr. Blake about one-eighth mile southeast of Brothers' locality number 4. This occurrence, although similar to others in the area, is characterized by an unusually fine development of lawsonite as veins and as lithologic layers parallel to the foliation of enclosing glaucophane schists. The locality is near the western edge of the estate, now the Blake Gardens, a research facility for the Department of Landscape Architecture of the University of California, and it and other outcrops of glaucophane schist on the grounds will be preserved.

Lawsonite and pumpellyite are found on the estate in a large block of glaucophane schist, 20 feet high and 25 feet across, apparently resting on serpentinite. The locality is probably within the Hayward fault zone, the main trace of which lies several hundred feet to the east. This schist block and others of similar or larger size in the area are regarded, in agreement with Brothers (1954, p. 616), as tectonic inclusions within serpentinites of the fault zone, although it is impossible to ascertain whether the Blake Gardens block is now *in situ*. Several other outcrops of serpentinite and glaucophane schist on the estate will not be described in this paper.

Occurrences in North Berkeley of lawsonite in tectonic inclusions of glaucophane schist are somewhat similar to the occurrence of this mineral in chlorite schist inclusions in serpentinite of Santa Clara province, Cuba (Schürmann, 1936). Exogenic blocks of eclogite and actinolite schist are found in the serpentinites of both areas. Glaucophane is not found in the lawsonite-bearing rocks of the Cuban association, but it does occur in some of the eclogites (Schürmann, 1936, p. 246).

GLAUCOPHANE SCHISTS

The glaucophane schists of the lawsonite-bearing block are fine-grained and have a well-defined foliation. They comprise three mineral associations:

1) glaucophane-sphene-(chlorite)-(colorless mica)
2) glaucophane-lawsonite-(sphene)
3) glaucophane-epidote-sphene

Anhedral apatite and spheroids of pyrite up to 2 cm in diameter are common accessory minerals of all associations. Small, ragged, often turbid grains of a pale green pyroxene believed to be relict are found in some specimens of the glaucophane-sphene and glaucophane-lawsonite assemblages.

The relict pyroxene occurs as individual grains and as aggregates parallel to the schistosity. It is consistently associated with a colorless mica, probably muscovite ($2V_x = 37°$-$44°$), and a pale green chlorite. An X-ray powder pattern of this mineral prepared by Dr. A. Pabst is identical with that of an acmitic diopside-jadeite from Healdsburg, California. The optics of this pyroxene are within the ranges described by Borg (1956, p. 1571) for the Healdsburg mineral: $2V_x = 76°$-$82°$ (Borg, 70°-82°) and $c\wedge Z = 49° \pm 1°$ (Borg, 44°-50°). Both Brothers (1954) and Borg (1956) consider glaucophane schists containing relict pyroxenes of this type to have been derived retrogressively from eclogite. Garnets were not found in schists of the pyroxene-bearing associations.

Glaucophane crystals commonly show color zoning, the intensity of color increasing outward in distinct zones from the crystal core. Typical pleochroic values are X = colorless to pale yellow, Y = violet to dark violet, and Z = sky blue to indigo blue. Universal stage measurements in sodium light give $2V_x$ variable between 35° and 45°. Dispersion is strong, $r < v$.

The epidote mineral pistacite occurs in amounts up to 25 percent by volume in some of the glaucophane schists. It is found as individual subhedral crystals and less commonly in aggregates of grains around which the foliation is deflected. The cores of most crystals are pleochroic in shades of green con-

forming to the pleochroic scheme X = colorless, Z = colorless to very pale apple green, and Y = pale apple green. Optic angle is negative and varies between 68° and 74°. Sections cut at high angles to an optic axis exhibit anomalous blue and yellow interference colors; dispersion r>v is moderate. The pistacite crystals, elongated parallel to Y, are often simply twinned on the composition plane {100}.

Anhedral to subhedral sphene grains are present in quantities up to 10 percent by volume in all of the glaucophane schists examined. These grains have a maximum size of 1 mm and are dichroic from tan to light brown; a small percentage exhibit elbow-shaped contact twins.

Lawsonite-bearing glaucophane schists (association 2) appear to be a variant of the first association resulting from crystallization of lawsonite aggregates along surfaces of schistosity. Alternation of light and dark-colored layers in the resulting rocks gives them a distinctly gneissic appearance. Lawsonite was not observed as discrete rock-forming grains within the glaucophane schists of this locality. The lawsonite-rich folia, discontinuous in lateral extent and 0.1 to 3.0 cm thick, are regarded as products of either metamorphic segregation or hydrothermal deposition, rather than a reflection of initial compositional layering. A total of 1450 point counts across a lawsonite-rich layer 1.3 cm wide gives the following mode computed in weight percent:

lawsonite	71.3	pyroxene	1.3
glaucophane	19.7	muscovite	0.1
pyrite	6.2	apatite	0.1
sphene	1.3		100.0

The above mode is considered representative of such layers, which are coarse-grained enough to permit megascopic comparison of the proportions of white lawsonite and blue-black glaucophane grains within them. From this mode an approximate chemical composition (table 1) for the lawsonite-rich band can be calculated by using chemical analyses of Berkeley Hills glaucophane and lawsonite (this paper). Pyrite is not included within this computation as it appears to be a late-stage post-lawsonite mineral.

TABLE 1

Calculated chemical composition of lawsonite-glaucophane layer in glaucophane schist, Blake Gardens

SiO_2	41.9
TiO_2	0.9
Al_2O_3	25.5
Fe_2O_3	2.8
FeO	2.6
MnO	0.1
MgO	1.6
CaO	14.3
Na_2O	1.5
K_2O	0.2
H_2O	8.6
	100.0

It is unlikely that the chemical composition of table 1 with its low silica, high lime-alumina content represents that of an original layer in the premetamorphic rock. As these glaucophane-lawsonite layers are cut by pumpellyite and lawsonite-pumpellyite veins of the variety to be described, their origin is probably best explained by a process of metamorphic differentiation occurring prior to the formation of the veins. It seems likely that the veins themselves are products of such a differentiation, perhaps a more mobile, aqueous culmination of the process giving rise to the lawsonite-glaucophane "gneisses".

VEIN MINERALS

The glaucophane schists just described are host to several generations of the vein minerals lawsonite, pumpellyite, and calcite; albite veinlets are present, some containing vugs lined with small euhedral crystals, but they are not common. With one notable exception veins are 0.2 to 1.5 cm wide and can be grouped into two sets governed by cross-cutting joints. An early set contains pumpellyite only or pumpellyite as the predominant vein mineral. These veins are consistently crosscut by a second set perpendicular to them in which lawsonite is generally the predominant mineral. Both sets contain calcite as the last mineral to be deposited. Textural evidence indicates deposition of the minerals in open spaces and by limited replacement of earlier vein minerals.

The exception noted above is a large lens-shaped vein, predominantly lawsonite, not related to or found cross-cutting either of the two joint sets described. This vein is unusual in its dimensions (maximum width 9 inches) and the large size of certain lawsonite crystals within it (2 to 3 inches in length). Lawsonite makes up most of the central portion of the vein and is bordered by a discontinuous pumpellyite zone up to 4 cm wide; pumpellyite also occurs as irregular patches within the lawsonite core. Several fragments of glaucophane schist completely enclosed by lawsonite and pumpellyite were found within the vein.

Lawsonite.—Lawsonite from the large vein is seen in thin section as anhedral crystals generally within the size range 0.2 to 2.0 cm. These crystals, tabular parallel to {010}, have maximum elongation parallel to c (=X). They possess perfect {010}, well-developed but discontinuous {100}, and imperfect {101} cleavages. The larger crystals are invariably strained and exhibit marked undulatory extinction and polygonization of the crystal into smaller, discretely bounded areas of variable extinction (1 to 20° variance within single crystals). Polysynthetic {101} and {$\bar{1}$01} twins are well developed only in those crystals lacking pronounced strain effects (X$\wedge${101} = 34°). The Blake estate lawsonite is slightly altered to a dusty unidentified phase spatially related to lawsonite cleavages. Calcite is associated with lawsonite as cavity fillings between anhedral-subhedral crystals, as veinlets cross-cutting lawsonite, and as minor localized patches replacing that mineral. Pumpellyite and lawsonite are frequently intergrown so that X of lawsonite is parallel to Y (=b) of pumpellyite (plate 1, fig. 1). This relationship has been previously described by Coombs (1953, p. 120) in terms of close correspondence between the a and b cell dimensions of lawsonite and pumpellyite, and consequent parallelism in intergrowth of their a and b axes ($a_L \parallel a_P$ and $b_L \parallel b_P$, or in the settings

PLATE 1

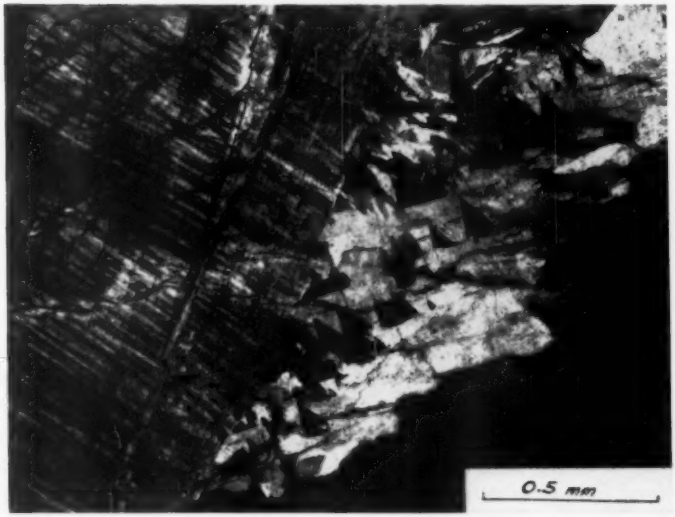


Fig. 1. Scalene-shaped intergrowths of pumpellyite (black) in lawsonite. Lawsonite with polysynthetic twinning on left. Crossed nicols.

Fig. 2. Sheath of pumpellyite crystals in upper center of picture with "oak-leaf twins" below. Crossed nicols.

adopted in this paper $a_L \parallel a_P$ and $c_L \parallel b_P$). The sequential relationships between lawsonite and pumpellyite are not always clear; in some cases, however, the intergrowth cited above is due to replacement of lawsonite by pumpellyite.

Optical properties of vein lawsonite from the Blake Gardens are summarized in table 2. These values agree quite closely with optical data on lawsonite from other localities.

TABLE 2

Optical properties, lawsonite, Blake Gardens

Orientation: X=c, Y=a, Z=b	
n_X	1.665 ± .002
n_Y	1.673 ± .002
n_Z	1.686 ± .002
n_Z-n_X	0.021 ± .004
$2V_{zNa}$	79-85°
Disp.	r>v, weak
Sp.G.	3.07 ± 0.01

A comparison of four superior chemical analyses of lawsonite, including an analysis of the Blake Gardens mineral, is presented in table 3. Formulae calculated from these analyses to conform to the simplified formula $CaAl_2Si_2O_8 \cdot 2H_2O$[1] illustrate extremely limited ionic substitution within the lawsonite structure.

Intergrown pumpellyite crystals are usually in the shape of a scalene triangle with interfacial angles, as measured by universal stage on a true {010} section of 97°, 128° and 135°. These angles correspond closely with calculated interfacial angles given by Coombs (1953, p. 117) for (001) Λ (100), (001) Λ (102), and (102) Λ (100) respectively and support his contention that the bounding faces for such scalene-shaped intergrowths are {001}, {100}, and {102} forms (1953, p. 127). However, variability of interfacial angle between the forms {100}? and {102}? noted by Coombs was also found in pumpellyite from the Blake Gardens, and over the same range (135-140°). Furthermore the interfacial angle between (001) and the face believed to be (100) is more consistently on the order of 92-93° than the 97° measured in one true {010} section. No reasons for such interfacial variability other than those suggested by Coombs (1953, p. 128) are offered here.

The scalene-shaped intergrowths may occur as isolated individuals (plate 1, fig. 1) or as series of back-to-back crystals usually separated by a narrow septum of lawsonite intergrown as previously described. The term "oak-leaf twin" has been applied by Coombs to back-to-back individuals in which {001} is a plane of symmetry between opposite crystals (plate 2, fig. 2). In some instances, however, back-to-back crystals in specimens from the Blake Gardens are not in twin relationship to each other, i.e. the crystallographic orientation in opposed individuals on either side of a lawsonite septum is the same.

Specific gravity of the Blake Gardens pumpellyite is 3.16 ± 0.01. Optical properties, in summary, are: $n_{yNa} = 1.689$; $2V_{zNa} = 51°\text{-}52°$; $Z \wedge c = 8°$; X

[1] Structural analysis by Wickman (1947) gives the formula $CaAl_2Si_2O_7(OH)_2 \cdot H_2O$.

and Z = colorless, Y = bright apple green; dispersion r<v is strong. Coombs (1953, p. 130-131) has demonstrated that the optical properties are correlative with iron content. Plottings of n_y, $2V_z$, and Z∧c against total iron content of 5.4 percent as Fe_2O_3 (see table 4) give points which fall on total iron-optics curves presented by Coombs.

Pumpellyite.—In the Blake Gardens, pumpellyite in the glaucophane schists examined is restricted to veins, although it is found as a rock-forming mineral elsewhere in the North Berkeley Hills (Brothers, 1954, p. 619). It is found as either bundle-like or matted aggregates of bladed crystals up to 2 cm long (plate 1, fig. 2), often with calcite or chlorite filling crystal interstices (plate 2, fig. 1), or as intergrowths with lawsonite as described above.

TABLE 3

Chemical analyses of lawsonite with corresponding atomic ratios

	A.	B.	C.	D.
SiO_2	38.14	38.81	37.80	38.45
Al_2O_3	30.91	32.02	32.56	31.35
Fe_2O_3	1.56	0.21	0.05	0.86
TiO_2	0.29	0.12	0.69	0.38
FeO	0.13	0.28	0.34	0.10
MnO	0.01	tr.	tr.	tr.
MgO	0.03	0.16	0.32	0.17
CaO	17.54	17.81	16.10	17.52
Na_2O	0.06	0.18	0.51	0.06
K_2O	0.14	0.21	0.24	0.23
H_2O	10.72	10.67	11.26	11.21
H_2O	0.00	0.36	0.04	
F_2	—	—	0.02	—
	99.53*	100.83	99.93	100.33

* Total loss on ign. = 11.40; theoretical H_2O_{total} = 11.42.

Atomic ratios calculated on basis of O = 8.

Si	2.014		2.009		1.978		2.008	
Al	1.914		1.954		2.007		1.929	
Fe'''	0.062	1.987	0.008	1.966	0.002	2.036	0.034	1.978
Ti	0.011		0.004		0.027		0.015	
Fe''	0.006		0.011		0.015		0.004	
		0.008		0.023		0.040		0.017
Mg	0.002		0.012		0.025		0.013	
Ca	0.988		0.988		0.902		0.980	
Na	0.006	1.003	0.018	1.019	0.052	0.970	0.006	0.997
K	0.009		0.013		0.016		0.011	
H_2O	1.887		1.844		1.964		1.946	

A. $(Ca_{0.988}Na_{0.006}K_{0.009})(Fe''_{0.006}Mg_{0.002})(Al_{1.914}Fe'''_{0.062}Ti_{0.011})Si_{2.014}O_8 \cdot 1.887\ H_2O$

B. $(Ca_{0.988}Na_{0.018}K_{0.013})(Fe''_{0.011}Mg_{0.012})(Al_{1.954}Fe'''_{0.008}Ti_{0.004})Si_{2.008}O_8 \cdot 1.884\ H_2O$

C. $(Ca_{0.902}Na_{0.052}K_{0.016})(Fe''_{0.015}Mg_{0.025})(Al_{2.007}Fe'''_{0.002}Ti_{0.027})Si_{1.978}O_8 \cdot 1.964\ H_2O$

D. $(Ca_{0.980}Na_{0.006}K_{0.011})(Fe''_{0.004}Mg_{0.013})(Al_{1.929}Fe'''_{0.034}Ti_{0.015})Si_{2.008}O_8 \cdot 1.946\ H_2O$

A. Blake Gardens, North Berkeley Hills, California. Doris Thaemlitz, anal.

B. Kanto Mountainland, Japan. Seki, 1957, p. 372. Y. Seki, anal.

C. Santa Clara province, Cuba. Schürmann, 1936, p. 250. van Tongeren, anal.

D. Tiburon Peninsula, Marin Co., California. Schaller and Hillebrand, 1904, p. 197. W. T. Schaller, anal.

PLATE 2

Fig. 1. Bladed pumpellyite crystals with interstitial chlorite.

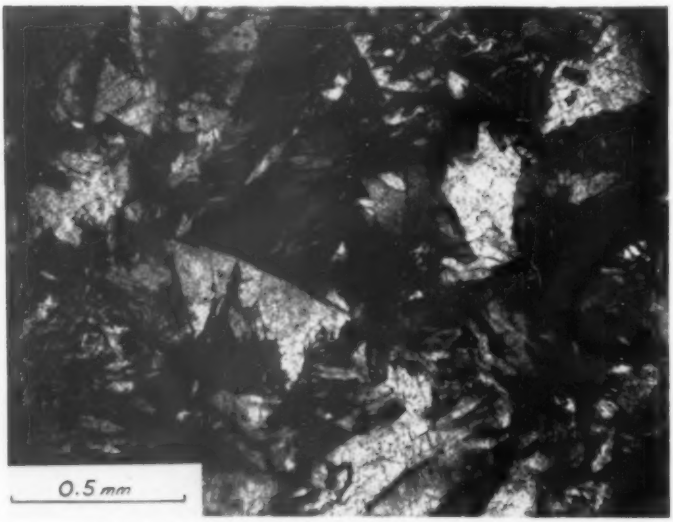


Fig. 2. "Oak-leaf twin" in pumpellyite. Crossed nicols.

TABLE 4

Semi-quantitative spectroscopic analysis, pumpellyite, Blake Gardens

Principal constituents: Ca, Al, Si

Fe_2O_3	5.4% *	Na_2O	0.1-0.2
TiO_2	0.5	V_2O_5	0.1
Cr_2O_3	0.01	Ni	0.002
MgO	5.0	Ga	0.005
MnO_2	0.1-0.5		

K, Zr, Cu, Ge, and Sr below level of sensitivity

* Quantitative determination, total iron expressed as Fe_2O_3. Average of two (5.1 and 5.7%). Analyst, George M. Gordon

Pumpellyite crystals in specimens from the Blake Gardens are elongated parallel to *b* (=Y) and are sometimes tabular parallel to {100}. Some crystals, diamond-shaped in {010} section, appear to be bounded by poorly developed {100} and {102} or {102} forms. Strong dispersion of the optic axes ($2V_{z_r} = 51°$-$52°$, $2V_{z_v} = 57°$-$59°$) results in anomalous blue and brown interference colors in sections cut at high angles to the optic plane.

DISCUSSION

Field relationships of glaucophane schist in the Blake Gardens and elsewhere in the North Berkeley Hills impose restrictions on petrogenetic interpretations pertaining to the schists themselves and the lawsonite and pumpellyite within them. The reader is referred to the paper by Brothers (1954) and a discussion of it by de Roever (1955) for opposing views on the origin of these schists. The present study, confined to the tectonic block on the western border of the Blake Gardens, suggests: 1) derivation of at least part of the glaucophane schists from eclogite; 2) formation of layered lawsonite-glaucophane rock by metamorphic differentiation; and 3) an alternating sequence of lawsonite and pumpellyite deposition in dilation fractures, these minerals being deposited from solutions derived by metamorphic differentiation accompanying glaucophanitic metamorphism.

ACKNOWLEDGMENTS

The writer would like to thank Professors A. Pabst and F. J. Turner for their interest in this project and for suggesting improvements in the manuscript. He is grateful to the Department of Geology of the University of California for funds for chemical and spectroscopic analyses. The late Mr. Anson Blake, a lifelong friend of Andrew C. Lawson for whom lawsonite was named, graciously permitted the writer to study the outcrops on his property. R. B. Parker is thanked for his assistance in preparing the photomicrographs.

NOTES ON THE X-RAY CRYSTALLOGRAPHY OF LAWSONITE

MATERIAL

Lawsonites from four localities, showing a variety of habits and associations, have been examined by single crystal and powder X-ray diffraction.

X-ray observations were also carried out on single crystals and powder after heat treatment. The following lawsonites were used:

1. from the type locality, Tiburon Peninsula, Marin County, California.
   (Ransome, 1895 and Schaller and Hillebrand, 1904)
2. from the Blake Gardens, Contra Costa County, California.
   (Occurrence described above by Mr. Davis)
3. from veins and cracks in low-grade metamorphic rock about 2½ miles northeasterly from Valley Ford, Sonoma County, California.
   (Some of the Franciscan rocks in this area have been described by Bloxam (1956) and the geology has been mapped by Travis (1952).)
4. from the Province of Santa Clara, Cuba.
   (Described by Schürmann (1936). Specimen kindly furnished by Dr. Schürmann.)

SETTINGS

Various settings have been used for lawsonite. The original setting chosen by Ransome (1895) was followed by Schaller and Hillebrand (1904) and by Rogers (1915). Gossner and Mussgnug (1931), who were the first to report X-ray observations on lawsonite, retained Ransome's orientation but found it necessary to double the *c* axis. Strunz (1937) chose a new orientation but adopted yet another setting in his *Mineralogische Tabellen* (1941 and later editions). This later Strunz setting was followed by Wickman (1947) who summarized the relations of the several settings in connection with his determination of the structure of lawsonite and by Seki (1957). Table 5 gives the transformations between all of the settings. In this report the setting chosen by Strunz in 1937 will be used. It conforms to the convention $c<a<b$ and has been used by Donnay (1951) and by Switzer (1951) who presented an angle table for lawsonite with indices referred to this setting.

TABLE 5

Lawsonite transformations

from \ to	Ransome 1894	Gossner & Mussgnug 1931	Strunz 1937	Strunz 1941
Ransome 1894		100/010/002	010/002/100	010/$\bar{1}$00/002
Gossner & Mussgnug 1931	100/010/00½		010/001/100	010/100/001
Strunz 1937	001/100/0½0	001/100/010		100/001/0$\bar{1}$0
Strunz 1941	010/$\bar{1}$00/00½	010/100/001	100/001/0$\bar{1}$0	

HABIT

The habit of lawsonite from the type locality has been pictured by Ransome (1895) and by Schaller and Hillebrand (1904). All crystals from this source examined during the current study were found to be short prismatic or nearly equidimensional, {101} usually being the dominant form. This is in contrast to the habit of lawsonite in the other specimens examined. In all of these the crystals, though sometimes in compact masses and sheaflike aggregates, are invariably elongated in the direction of the *c* axis (*b* axis of Wickman) as would be expected from the cell dimensions and structures. About a dozen crystals were measured. All faces observed are in the zone of the *c* axis. Generally a near continuum of reflections is observed over much of the *hk*0 zone but no indices other than those of forms listed by Switzer, {100}, {010} and {210}, could be surely established. No crystals with *b* axis elongation such as shown by Switzer (1951, fig. 2) were encountered.

CELL DIMENSIONS

The cell dimensions of lawsonite from the four localities were determined by means of quartz-calibrated Weissenberg patterns. All results are shown in table 6. It is clear that any variations in the dimensions are very slight. The average values with the stated limits can be considered to apply to any of these lawsonites. In the lower part of table 6 are given the cell dimensions previously reported for lawsonite. Wickman (1947) examined lawsonite from the type locality and it may be assumed that Gossner and Mussgnug (1931), who did not state the provenance of their material, obtained it from the same source. The marked differences in the dimensions for lawsonite reported by these observers from the newly determined dimensions are puzzling. Seki (1957) examined lawsonite from the Kanto Mountainland, Japan. The small differences between the cell dimensions he reported and those newly determined may be due to real differences in the materials.

TABLE 6

Cell dimensions of lawsonite

	a_o	b_o	c_o
Tiburon	8.786		5.848
xl. #2		13.134	5.851
Blake Gardens	8.787	13.127	
xl. #5		13.127	5.842
Valley Ford	8.793		5.848
xl. #3	8.786	13.134	
Cuba		13.127	5.842
xl. #3	8.785	13.126	
average	8.788 ± 0.008	13.129 ± 0.010	5.846 ± 0.006 Å
Gossner & Mussgnug (1931)	8.87	13.25	5.88 Å *
Wickman (1947)	8.90	13.33	5.76 Å *
Seki (1957)	8.75	13.09	5.84 Å

* Converted to Å units from the values originally reported in kX units.

POWDER PATTERN

The only published records of X-ray powder diffraction patterns of lawsonite known to the writer are that of Seki (1957) with observations in terms of 2θ and ASTM card 8-137. In table 7 is given the record of concurrent measurements, over the same angular range as given by Seki, on powder patterns of the three California lawsonites examined, presented in the conventional manner in terms of d-spacings and indexed to conform to the setting here chosen. To convert to the indexing of Seki, which is also given, k and l must be interchanged. Intensities were estimated by superposition of a scale of spots of graded intensities. Except for minor differences in resolution and the recording of some very weak reflections the agreement with Seki's report is excellent. However, Seki indexed lines at 2θ 31.36° and 37.04°, equivalent to spacings 2.852 and 2.429, 021 and 023 respectively. This is permissible in the space group $C222_1$ chosen by Wickman, but lawsonite shows false symmetry due to the special values of parameters of all atoms in general positions (Pabst, 1959) which results in zero intensity for reflections of this type. The space group symbol in the new orientation is $B22_12$ and the symbol of the space group simulated due to the false symmetry is $B\ 2/b\ 2_1/m\ 2/m$. The indexing has been checked for every recorded line by comparison with precession or Weissenberg patterns. (See postscript.)

HEATED LAWSONITE

A few yards from the lawsonite outcrops in the Blake Gardens some rock fragments resembling lawsonite vein material but brownish in color were found in a spot where there was evidence of old bonfires. A small crystal selected from such a specimen was examined by means of an oscillation pattern. This showed the normal diffraction spots for lawsonite plus a great number of strong powder arcs which were readily identified as due to the metastable hexagonal modification of $CaAl_2Si_2O_8$ (Davis and Tuttle, 1952). Upon heating this partly changed crystal to 760°C it was completely converted to hexagonal $CaAl_2Si_2O_8$. Goldsmith and Ehlers (1952) have reported that this phase is produced when lawsonite is heated in air for 24 hours at 650°C, that anorthite begins to form after 24 hours at 800°C and that the conversion can be completed at 1,050°C in 4 hours or less. Similar results were obtained by Douglass (1954). Goldsmith and Ehlers stated that "the lowest temperature at which lawsonite decomposes in air in a reasonable time has not yet been established." Switzer (1951, fig. 3) published a differential thermal curve for lawsonite and stated "A large endothermic peak at 641°C and a smaller one at 744° indicate loss of water at two different temperatures." A differential thermal curve of lawsonite from the Blake Gardens, kindly run by Mr. Larry Godwin, shows similar features.

TABLE 7

X-ray powder diffraction pattern of lawsonite

$c<a<b$ hkl	$b<a<c$ hkl	$d_{calc.}$	$d_{obs.}$*	I	$c<a<b$ hkl	$b<a<c$ hkl	$d_{calc.}$	$d_{obs.}$*	I
020	002	6.56	6.58	3½	113	131	1.883	1.884	1½
101	110	4.87	4.84	6	351	315	1.854	1.852	½
111	111	4.56	4.57	3	123	132	1.827		
200	200	4.39		3	440	404	1.826		
210	201	4.17	4.18	4	252	225	1.785	1.786	½–
121	112	3.91	3.91	½	402	420	1.756		
220	202	3.65	3.66	6	062	026	1.752	1.752	3½
040	004	3.28			171	117	1.750		
			3.26	3½	133	133	1.745		
131	113	3.25			412	421	1.741		
230	203	3.10			270	207	1.725	1.726	2
002	020	2.923	2.925	4½	422	422	1.696	1.697	1½
141	114	2.721	2.726	7	450	405	1.685	1.685	½
022	022	2.670	2.680	5	501	510	1.683		
240	204	2.629			361	316	1.679		
			2.624	10	511	511	1.669		
301	310	2.619		10				1.657	½
311	311	2.579	2.573	½	143	134	1.646		
202	220	2.434			080	008	1.641		
			2.433	6	521	512	1.630	1.631	3
321	312	2.432			432	423	1.630		
212	221	2.393	2.395	3	262	226	1.627		
151	115	2.311	2.311	1½				1.626	4
222	222	2.282	2.284	2	303	330	1.622		
250	205	2.254	2.254	3½	313	331	1.610		
331	313	2.247			323	332	1.575	1.575	1–
400	400	2.197			531	513	1.571		
			2.190	1½B	181	118	1.555		
060	006	2.188			460	406	1.550	1.550	8
042	024	2.182			442	424	1.548		
410	401	2.167			153	135	1.541		
232	223	2.127	2.129	6	280	208	1.537		
420	402	2.083	2.089	½	371	317	1.525		
341	314	2.047	2.053	½	333	333	1.521		
161	116	1.996	1.990	½	541	514	1.499	1.501	1
430	403	1.963	1.965	3	272	227	1.486		
260	206	1.959			600	600	1.483		
242	224	1.955			004	040	1.461	1.464	4
103	130	1.902			452	425	1.460		

* Cu radiation, Ni filter; Camera diameter 114.59 mm, Straumanis setting, Measurements corrected for film shrinkage.

Some observations were made on the sequence of changes when lawsonite is heated in air. Single crystals were heated to known temperatures for various periods and then examined by *c* axis oscillation patterns after each period of heating. Five crystals from the Blake Gardens and one from Cuba, all about 0.5 x 0.1 x 0.1 mm, were used.

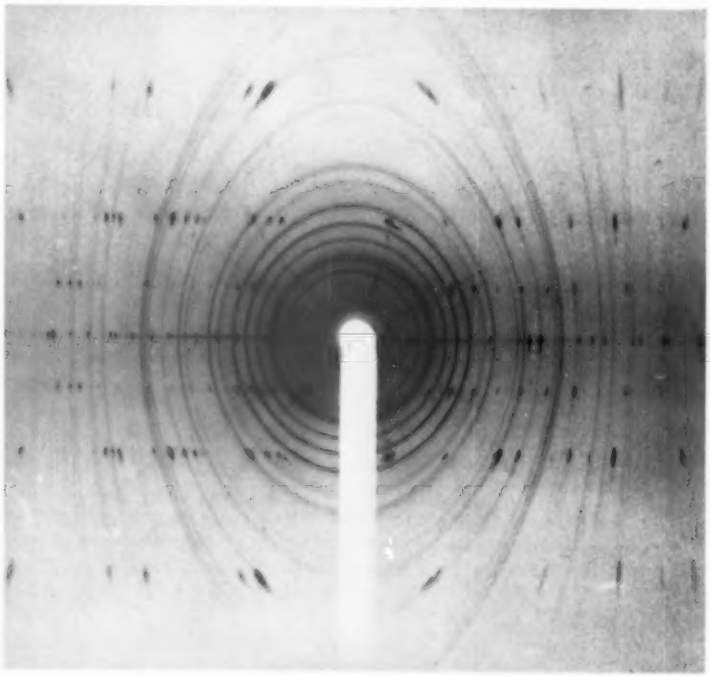
Besides the powder lines of hexagonal $CaAl_2Si_2O_8$ a few of the patterns of heated lawsonite show a single very weak line near 3.2 Å. The strongest line of orthorhombic $CaAl_2Si_2O_8$ is at 3.21 (Davis and Tuttle, 1952) and the strong-

est anorthite line at 3.20 Å. Since anorthite is formed only at higher temperatures (Goldsmith and Ehlers, 1952, p. 395) it seems reasonable to attribute this line to a minute amount of orthorhombic $CaAl_2Si_2O_8$ in those cases where it appears in patterns obtained from crystals heated to 500°C or less. In the one case in which it appeared in a pattern of material heated to 700°C, it might be attributed to anorthite. Some of the patterns of material heated for very long periods or at rather high temperatures (670 to 760°C) show a weak, diffuse line near 3.35, close to the position of the strongest quartz line, but one would hardly expect the appearance of quartz and so this line remains unexplained.

Heatings were carried out for periods from half an hour to five days and at temperatures from 410° to 760°C. The results may be summarized as follows:

A. At 410°C. After 16 hours no change can be observed in a single crystal by oscillation pattern. After 40 hours the strongest powder arcs due to hexagonal $CaAl_2Si_2O_8$ can be seen.

PLATE 3



c axis oscillation pattern of lawsonite crystal from Blake Gardens after heating for 154 hours at 450°C. Cu radiation, Ni filter, 6-hour exposure.

B. At 450°C. After 3 hours no change is discernible in a single crystal. After 16 hours a few powder arcs are superposed on a sharp single crystal pattern. Lawsonite ground to less than 300 mesh and heated for 16 hours at this temperature is not sufficiently transformed to allow detection by powder pattern. After heating the powder for a total of 34 hours at this temperature the three strongest lines of the dehydration product are discernible in the pattern among the full complement of lawsonite lines. A single crystal heated at this temperature in several increments for a total of 154 hours yields the complete hexagonal $CaAl_2Si_2O_8$ powder pattern superposed on a lawsonite oscillation pattern of sharp spots with intensity scarcely diminished (pl. 3).

C. At 555°C. A crystal heated at 3 hours to this temperature after previous heatings of 3 hours at 455° and 15 plus 21 hours at 515°C shows only the last traces of the lawsonite single crystal pattern with the hexagonal $CaAl_2Si_2O_8$ pattern but the few remaining spots are sharp, though weak.

D. At 580°C. The same crystal was completely converted by further heating for 16 hours.

E. At 690°C. A single crystal after 35 minutes yields a complete, very sharp, hexagonal $CaAl_2Si_2O_8$ powder pattern and only a few weaks spots of a lawsonite oscillation pattern. Powdered lawsonite after the same period of heating yields no trace of unchanged material by X-ray pattern.

F. At 700°C. A single crystal is completely converted in an hour and a half or less. The initially clear crystal is porcelaneous in appearance after conversion.

These observations show that the thermal decomposition of lawsonite to hexagonal $CaAl_2Si_2O_8$ begins very slowly at 410°C or less, at least 230° below the most prominent endothermic DTA peak at 641°C, and proceeds rapidly, being complete in approximately an hour, in the neighborhood of 700°C well below the second endothermic DTA peak at 744°C. The first stages of conversion can be recognized by oscillation patterns on single crystals when they are not detectable by powder pattern. Initially clear crystals show only the faintest milkiness in these stages. In the range from about 410 to about 550°C lawsonite crystals are converted very slowly or maybe only partly, the change being incomplete even after heating for several days.

Postscript (February 5, 1960).—After this paper had been accepted for publication our attention was drawn to a paper on "The crystalline structure of lawsonite" by I. M. Rumanova and T. I. Skipetrova (1959 Doklady, Akad. Nauk, S. S. S. R., v. 124, p. 324-327, [GeoScience Abstracts, abstract 1-3099, Dec. 1959 and Chem. Abst. 53, 9914*e*, June 10, 1959]). These authors report the cell dimensions $a = 8.83$ Å, $b = 5.80$, $c = 13.20$, using the setting of Strunz (1941) and Wickman. They arrive at a structure in the space group *Ccmm*, which requires the systematic absences that Pabst (1959) attributed to the false symmetry associated with Wickman's structure in the space group $C222_1$. See also "Considerations on false symmetry in the structure of lawsonite" by I. M. Rumanova and N. V. Belov, 1960, Kristallografiya, v. 5, p. 215-217.

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[American Journal of Science, Vol. 258, December 1960, P. 705-715]

JADEITE IN SANBAGAWA CRYSTALLINE SCHISTS OF CENTRAL JAPAN

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ABSTRACT. Jadeite is observed in pelitic, psammitic and mafic crystalline schists in a low-grade part of the Sanbagawa metamorphic belt of central Japan. Stable association of jadeite and quartz in some of these schists is believed to show that the temperature and pressure prevailing in the formation of these schists may have been 200-300°C and 6-12 kilobars respectively.

INTRODUCTION

Formerly, jadeite was regarded as a very rare mineral in the world, and the details on its modes of occurrence were not known. Within the last ten years, however, it has become clear that jadeite is widespread in some low-grade crystalline schists of Celebes, California and Japan (de Roever, 1955; Bloxam, 1956, 1959; McKee, 1958; Seki, Aiba and Kato, 1960; Seki and Shidô, 1959; Shidô and Seki, 1959). These crystalline schists are of glaucophanitic type. Metamorphic minerals associated with jadeite are lawsonite, pumpellyite, epidote, glaucophane, chorite, white mica, sodic plagioclase, quartz and others.

Synthetic and calorimetric works on jadeite have proved that the formation of jadeite requires high pressures, especially when quartz is associated with it.

In spite of this recent progress, an important problem has not yet been solved. It is in what grade or subfacies of glaucophanitic metamorphism jadeite is stable. In order to solve this problem, the distribution of jadeite must be investigated in relation to zonal mapping of glaucophanitic metamorphic terrains. A brief historical review of the zonal mapping and subfacies in glaucophanitic metamorphism will be given as an introduction.

De Roever (1947 and 1950) was the first to propose subdivision of the glaucophane schist facies. In his study of the regional metamorphic rocks of Celebes, he subdivided them into three subfacies: epidote-garnet-glaucophane-, lawsonite-garnet-glaucophane-, and lawsonite-glaucophane subfacies in the order of descending grade of metamorphism. In 1955, he reported the occurrence of jadeite in some rocks of his lawsonite-glaucophane subfacies. Since his works were confined to study of specimens collected by other workers and no detailed field work was made in combination with his mineralogical study, his subfacies were open to criticism.

Brouwer and Egeler (1952) made a petrological study on the glaucophanitic regional metamorphic rocks in the eastern part of Corsica. They suspected the possibility of subdivision of this terrain into the following three zones from north to south: epidote-glaucophane zone (garnet is stable, but lawsonite is unstable), garnet-lawsonite-glaucophane zone (epidote is unstable) and lawsonite-glaucophane zone (epidote and garnet are unstable). The first zone would show the highest grade of metamorphism and the last one appear to show a transitional relation to rocks almost or entirely devoid of metamorphic features. Jadeite was not found from the Corsica terrain,

though they found a common occurrence of "green soda-pyroxene", which is probably rich in aegirine molecule judging from the optical data. No geological map showing zonal distribution of rocks or minerals was presented. Thus their subdivision could not be accepted unreservedly.

Routhier (1953) reported some interesting facts on the repartition and stability of lawsonite and epidote in glaucophanitic metamorphic terrain of New Caledonia. These facts are in good agreement with others discovered in Celebes and in Corsica by de Roever, and Brouwer and Egeler.

Bloxam (1956) found that jadeite is a common mineral in greywackes in a part of California. The jadeite is stably associated with quartz, sodic plagioclase, glaucophane and lawsonite. It was formed in greywackes probably by the reaction albite → jadeite + quartz under low-grade metamorphic conditions, locally augmented by deformation. He stated "it is most likely that the jadeite-bearing greywackes may occur in other parts of California but have hitherto escaped notice because of their resemblance to unmetamorphosed greywacke". This statement was verified by the recent discovery, by McKee (1958), of an areal distribution of jadeite-bearing greywackes in the Franciscan series of California.

Borg (1956) described lawsonite-glaucophane schists, eclogite and other kinds of interesting low-grade metamorphic rocks in the Healdburg district, California. Though she does not pay any special attention to the spatial distribution of rocks, it appears to the present writer that figure 2 in her paper (and also figure 1 in Bloxam's paper (1959)) may represent the possibility of subdividing the area into two or more zones of different metamorphic conditions.

Banno succeeded in zonal mapping of the glaucophanitic regional metamorphic terrain of the Omi district in central Japan (Banno, 1958; Miyashiro and Banno, 1958). He divided the area into two zones: chlorite zone and biotite zone. Glaucophane is stable only in the chlorite zone. Neither lawsonite nor jadeite has been found in the schists themselves, though jadeite occurs in inclusions within the associated serpentinites. Pumpellyite has not been found.

Recently, the present writer succeeded in zonal mapping of the Sanbagawa glaucophanitic regional metamorphic terrain of the Kantô Mountains of central Japan (Seki, 1958). He divided the terrain into six zones showing the progressive mineralogical changes: (1) unmetamorphosed zone, (2) pumpellyite-glaucophane-actinolite zone, (3) pumpellyite-epidote-glaucophane-actinolite zone, (4) lawsonite-pumpellyite-epidote-glaucophane-actinolite zone, (5) pumpellyite-epidote-glaucophane-actinolite zone, and (6) epidote-garnet-actinolite zone. The metamorphic conditions vary continuously through this series of zones. When the writer published this paper, he was not aware of the occurrence of jadeite in these metamorphic rocks.

Miyashiro and Seki (1958) proposed the following two subfacies of the glaucophane schist facies on the basis of the studies of the Omi district and the Kantô Mountains: epidote-glaucophane subfacies and lawsonite-pumpellyite-epidote-glaucophane subfacies. They stated that epidote-glaucophane subfacies represents higher solid pressures than the green schist facies, if the

temperature and water pressure may be regarded as the same, and lawsonite-pumpellyite-epidote-glaucophane subfacies represents lower temperature and/or higher water pressures than the epidote-glaucophane subfacies. They considered that jadeite is probably unstable in the lawsonite-pumpellyite-epidote-glaucophane subfacies, because jadeite was not found in any rocks of the Kantô Mountains before their paper was published.

In 1959, Seki and his co-workers succeeded in zonal mapping of the Sibukawa crystalline schist area of central Japan. This area is a part of the Sanbagawa metamorphic belt (Seki, Aiba and Kato, 1959a and 1959b). This district was divided into the following three zones: (1) pumpellyite-epidote-glaucophane-actinolite zone, (2) garnet-epidote-glaucophane-actinolite zone and (3) epidote-edenite zone. Within the second zone, there are some elongated areas in which lawsonite is stably present in association with pumpellyite, epidote, glaucophane and actinolite in crystalline schists. Jadeite, near the ideal composition, was also found in several meta-gabbroic rocks of the present district (Seki, Aiba and Kato, 1960). All of these jadeite-bearing meta-gabbroic rocks were found in serpentinites situated within the crystalline schists representing the mineral assemblages of the lawsonite-pumpellyite-epidote-glaucophane subfacies of Miyashiro and Seki (1958).

Immediately afterwards, Seki found jadeite in the Sanbagawa metamorphic rocks of the Kantô Mountains and the Sibukawa district, and Shidô and Seki also found jadeite in the Kamuikotan metamorphic belt of Hokkaido, north Japan (Seki and Shidô, 1959; Shidô and Seki, 1959). It was noticed that the crystalline schists in these jadeite-bearing areas appear to belong to lawsonite-pumpellyite-epidote-glaucophane subfacies. Though the Kamuikotan belt has been noted for the occurrence of glaucophane, aegirinaugite, lawsonite and pumpellyite, no systematic petrological study has been made. In this belt there are various grades of crystalline schists from epidote amphibolite to very low-grade metamorphic rocks. There is very good hope that the stable range of jadeite in this metamorphic terrain may be settled on the basis of detailed field, mineralogical and chemical work.

JADEITE IN THE SANBAGAWA METAMORPHIC TERRAINS OF THE KANTÔ MOUNTAINS AND SIBUKAWA DISTRICT, JAPAN

Identification of jadeite

The identification of jadeite in crystalline schists of the Sanbagawa belt is a hard task. Most of the jadeites are fine grained, that is 0.05-0.3mm in diameter, and are closely mixed with other metamorphic minerals such as lawsonite, pumpellyite, epidote, glaucophane, actinolite, chlorite, white mica, sodic plagioclase and quartz. Often it is very difficult to discriminate jadeite in thin sections from lawsonite, pumpellyite (especially Al-rich pumpellyite), actinolite and white mica.

During the present investigation, the present writer made optical examination of more than 2500 rocks. When we found a mineral which may be regarded as jadeite so far as the optical properties are concerned, we separated the mineral and examined it by the x-ray method. Thus, 98 samples were examined and it was found that 52 samples among them contain jadeite.

Optical properties and x-ray powder pattern of jadeite

Jadeite in the crystalline schists of the Kantô Mountains and the Sibukawa district have the following optical properties:

$$\left.\begin{array}{l}\alpha = 1.650\text{–}1.655\\ \beta = 1.657\text{–}1.661\\ \gamma = 1.667\text{–}1.673\end{array}\right\} \gamma\text{–}\alpha = 0.017\text{–}0.021.$$

(+) $2V = 63\text{–}75°$. $c\Lambda Z = 39°$.

b=Y. Colorless in thin sections. $r < v$.

Measurements of optical angle, optical orientation and extinction angle were made on ordinary thin sections by means of the polarizing microscope and universal stage for Na-light. Indices of refraction were measured by the immersion method using Na-light.

Jadeite separated from crystalline schists by means of the isodynamic separator and Clerici solution were examined by the Philips Geiger counter x-ray diffractometer using Cu-radiation. Three characteristic peaks at $2\theta = 31.5°$, $30.5°$ and $28.65°$ ($CuK\alpha$) in the powder pattern were used for identification (Yoder, 1950; Seki, Aiba and Kato, 1960).

Distribution of jadeite

Kantô Mountains.—The geology and petrology of the Sanbagawa metamorphic area of the Kantô Mountains have been already described in detail in another paper (Seki, 1958).

On the basis of the distribution of metamorphic minerals, the grade of recrystallization and stratigraphical sequences, the present area can be divided into six zones as stated on an earlier page.

Jadeite is stable only in the lawsonite-pumpellyite-epidote-glaucophane-actinolite zone. Original rocks of these jadeite-bearing crystalline schists can be easily determined because of the poorness in the degree of recrystallization. They are pelitic and psammitic sediments and mafic volcanic rocks. As will be mentioned in a later chapter, most of these jadeites are closely associated with lawsonite.

No special structural and tectonic differences were noticed between these jadeite-bearing rocks and other schists.

Sibukawa district.—The present writer and his students have already described the geology and metamorphism in the Sibukawa district, central Japan (Seki, Aiba and Kato, 1959a, 1959b and 1960).

As mentioned in an earlier page of this paper, this district can be divided into three zones representing progressive metamorphism. Jadeite was found in crystalline schists in elongated areas of lawsonite-pumpellyite-epidote-glaucophane subfacies located within the pumpellyite-epidote-glaucophane-actinolite zone. Jadeite-bearing meta-gabbroic rocks were also found enclosed in some ultramafic masses intruded into these lawsonite-bearing areas (Seki, Aiba and Kato, 1960). No remarkable structural and tectonic differences between jadeite-bearing schists and other crystalline ones was noticed.

Petrography of jadeite-bearing schists

There are two types of original rocks for jadeite-bearing crystalline schists

in the Kantô Mountains and Sibukawa district: mafic volcanic rocks on the one hand and pelitic and psammitic rocks on the other.

Jadeite-bearing schists derived from mafic volcanic rocks.—These crystalline schists are dark green, dark bluish green or greenish violet in color. The schistosity is weak or moderately distinct. Linear structure represented by the microfolding of schistosity and by the oriented arrangements of constituting minerals is also found in most of these rocks. Some pyroclastic structures can be easily recognized in hand-specimens or exposures of jadeite-bearing mafic schists.

Crystals of relict augite are common. They are partly replaced by colorless or pale green actinolite, glaucophane, chlorite and jadeite along their crystal margins or cleavages. Most of the augites show a moderate degree of wavy extinction.

Original calcic plagioclase was completely replaced by sodic plagioclase, jadeite, lawsonite, pumpellyite, epidote and/or chlorite.

Jadeite shows various modes of occurrences. Some of it has grown along the margin of relic augite, keeping the c-axis completely or nearly parallel to that of the augites. In other cases, jadeites having tabular, granular or, though rarely, fibrous forms are stably associated with chlorite, glaucophane, lawsonite, sodic plagioclase and/or epidote. Assemblages of jadeite, chlorite and other minerals form clusters, lenses or veinlets in some of the crystalline schists. Some of these jadeites commonly show characteristic clear polysynthetic twinning on (110).

One of the most significant minerals, closely associated with jadeite, is lawsonite. This mineral is always in tabular form and is present in most jadeite-bearing mafic schists. The lawsonite is stably associated with jadeite, glaucophane, chlorite, epidote, pumpellyite, actinolite, though the association of lawsonite with actinolite is rather rare.

Most jadeite-bearing mafic schists contain epidote, in a smaller amount than lawsonite. The epidote shows strong birefringence and comparatively small optical angle over X (67-82°). Most of these epidotes show zonal structure, having the margin of weaker birefringence and larger optical angle over X than the core.

The pumpellyite in jadeite-bearing mafic schists is usually associated with lawsonite and/or epidote. It is generally fibrous and shows radial arrangement. Most of these pumpellyites are pleochroic from pale green to colorless, or from yellowish green to colorless. Some pumpellyites, however, are colorless in all directions, and have low refractive indices. They are probably rich in Al and Mg. Such pumpellyites can hardly be discriminated from fibrous jadeite in thin sections. However a difference in refractive indices, optical orientation and X-ray powder patterns are diagnostic in the discrimination of these minerals. The lowest refractive index of pumpellyite is 1.670, while the highest value of refractive indices of jadeite is 1.668.

The glaucophane and actinolite associated with jadeite have the optical orientation of b = Y. The index of refraction β of the glaucophane is lower than 1.650. The highest value of index β of actinolite is 1.670. Both of these minerals show always tabular or fibrous forms.

Other important minerals of jadeite-bearing mafic schists are quartz and sodic plagioclase. Sodic plagioclase ($\alpha = 1.530 \pm 0.002$) was found in most of jadeite-bearing schists, stably associated with lawsonite, glaucophane, chlorite, epidote and white mica. Quartz occurs in some jadeite-bearing mafic schists, though the amount of this mineral is far less than that of sodic plagioclase. Some of jadeite-bearing mafic schists have neither sodic plagioclase nor quartz. The presence of lenticular or vein-like segregates chiefly composed of jadeite, lawsonite, sodic plagioclase and/or quartz in some jadeite-bearing mafic schists may represent the stable association of jadeite with sodic plagioclase and/or quartz.

Titanite, hematite, chlorite and white mica were also found in some jadeite-bearing mafic schists. No analcite was found.

Chemical compositions of some jadeite-bearing mafic crystalline schists are represented under columns 1 and 2 in table 1.

TABLE 1.

Chemical compositions of jadeite-bearing crystalline schists of the Kantô Mountains. These analyses were already represented in a previous paper by the present writer (Seki, 1958), though the presence of jadeite was not reported.

	YS54010207	F154011208	CK57050406
SiO_2	46.53	47.45	58.71
TiO_2	n.d.	n.d.	0.61
Al_2O_3	15.31	9.78	13.32
Fe_2O_3	20.53	6.21	3.36
FeO	2.00	6.27	7.69
MnO	n.d.	0.37	0.83
MgO	2.70	6.95	2.71
CaO	7.08	15.38	5.93
Na_2O	4.46	2.10	1.71
K_2O	0.26	0.28	0.92
H_2O+	1.77	5.36	4.61
H_2O-	0.34	0.73	0.33
Total	100.98	100.88	100.73

Jadeite-bearing pelitic and psammitic schists.—Jadeite-bearing schists derived from pelitic and psammitic rocks are black, gray or greenish-gray in color. They have distinct schistosity. These rocks can not be discriminated from other ordinary pelitic and psammitic schists in the field.

Metamorphic minerals of these jadeite-bearing pelitic and psammitic schists are as follows: jadeite, lawsonite, quartz, sodic plagioclase, chlorite, white mica, stilpnomelane, hematite, titanite and graphite. These minerals are believed to be stably associated with each other; at least, no unstable relation can be inferred from their modes of occurrence. The association of quartz-jadeite and quartz-jadeite-sodic plagioclase are common in these schists. Epidote, pumpellyite and amphibole-group minerals were not observed. The chemical composition of a jadeite-bearing pelitic schist is represented under column 3 in table 1.

Summary

The mineral assemblages of jadeite-bearing schists in the Sanbagawa metamorphic terrains of the Kantô Mountains and the Sibukawa district may be summarized in the A-C-F diagram as shown in figure 1. This diagram is the same as that given for the lawsonite-pumpellyite-epidote-glaucophane subfacies in glaucophane schist facies by Miyashiro and Seki (1958).

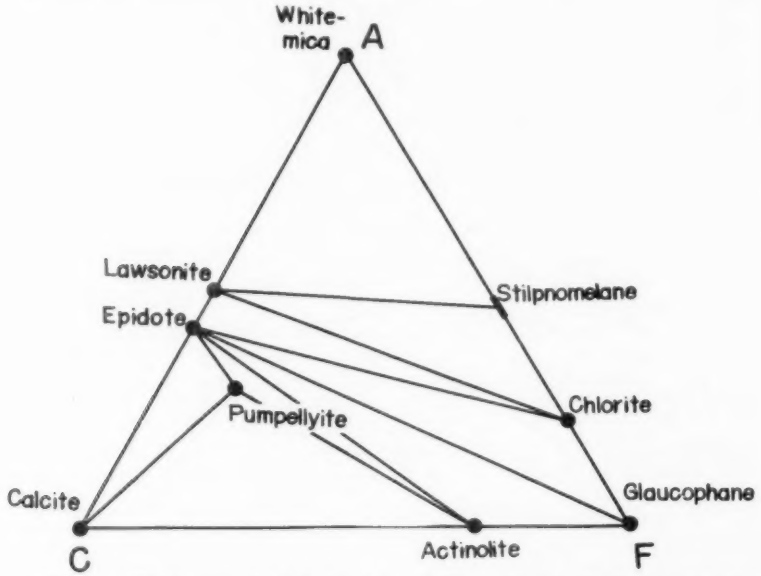

Fig. 1. ACF diagram of jadeite-bearing crystalline schists in the Sanbagawa metamorphic belt. This is the same as that of the lawsonite-pumpellyite-epidote-glaucophane subfacies of glaucophane schist facies (Miyashiro and Seki, 1958).

In figure 1, jadeite can not be represented. So the present writer tried to make another diagram: $(Al,Fe)_2O_3$–CaO–(Mg,Fe)O–Na_2O diagram (fig. 2). In this diagram, the $(Al,Fe)_2O_3$ corner represents the total amount of Al_2O_3 and Fe_2O_3 in the analysis. (It is to be noted that the quantity of Al_2O_3 that would be required to form feldspar, glaucophane and jadeite is not subtracted.)

As was already shown in table 1, the associations of jadeite with sodic plagioclase, lawsonite, epidote, glaucophane, actinolite and chlorite are common in the jadeite-bearing mafic schists. On the other hand, white mica is rather rare in these schists. In jadeite-bearing pelitic and psammitic schists, the mineral assemblages of jadeite, lawsonite, chlorite, sodic plagioclase, quartz and white mica are common. Glaucophane, actinolite, epidote, pumpellyite and calcite are not present.

Glaucophane and white mica are quite antipathetic to each other in occurrence. In most jadeite-bearing mafic schists with stable glaucophane, white mica is very rare. White mica, however, occurs, though not so common, in

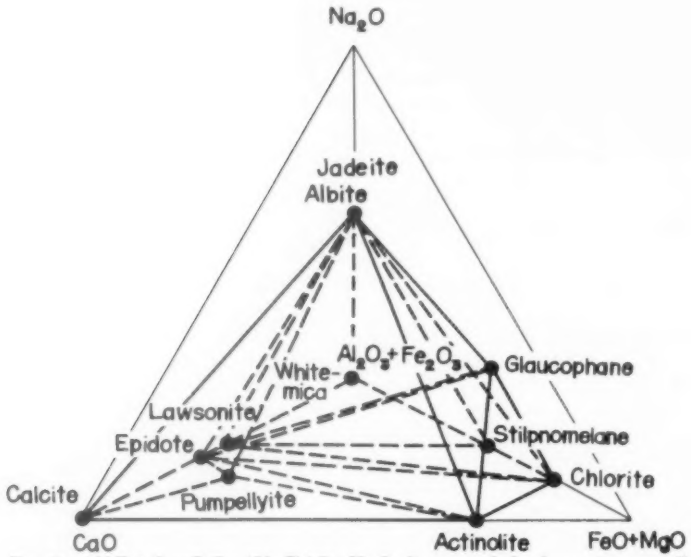

Fig. 2. $(Al,Fe)_2O_3$—CaO—(Mg,Fe)O—Na_2O diagram of the lawsonite-pumpellyite-epidote-glaucophane subfacies. $(Al,Fe)_2O_3$ was given by the total Al_2O_3 and Fe_2O_3, not less than the quantity that would be required to form feldspar, glaucophane and jadeite.

glaucophane-free jadeite-bearing mafic schists. In jadeite-bearing schists derived from pelitic and psammitic rocks, glaucophane is quite absent, while white mica is usually present.

These mineralogical features would be due to difference in chemical composition between mafic volcanic rocks and pelitic and psammitic sediments. The chemical compositions of mafic volcanic rocks would be generally represented by points within the three-dimensional field limited by jadeite, sodic plagioclase, lawsonite, epidote, actinolite, chlorite and glaucophane in figure 2. It is also very likely that the chemical composition of pelitic and psammitic sediments are generally represented by points within the field limited by jadeite, lawsonite, chlorite and white mica.

DISCUSSION

As described above, jadeite occurs in very limited parts within the Sanbagawa metamorphic belt. The mineral assemblages of rocks in these parts are shown in figures 1 and 2. These figures are believed to represent a definite mineral facies or subfacies, that is, lawsonite-pumpellyite-epidote-glaucophane subfacies of the glaucophane schist facies. Jadeite is one of the critical minerals of this subfacies.

Physical conditions of lawsonite-pumpellyite-epidote-glaucophane subfacies.—Judging from the constituent metamorphic minerals such as jadeite, lawsonite, pumpellyite, epidote, glaucophane, actinolite, chlorite, white mica and sodic plagioclase, the temperature of the lawsonite-pumpellyite-epidote-glaucophane subfacies is not high. The metamorphic rocks of the present sub-

facies represent a lower-grade part of the Kantô Mountains and Sibukawa district. In these districts, the mineralogical assemblages of schists in the highest-grade parts are very similar to those of the ordinary green schist facies. So, the present writer believes that the temperature of the lawsonite-pumpellyite-epidote-glaucophane subfacies with jadeite is rather low, say, below 300°C.

Lawsonite is very rich in water. Probably, high water pressure or high chemical potential of water during metamorphism is a necessary condition for the formation of lawsonite. Most of the other metamorphic minerals in the subfacies are also moderately rich in water. In a certain range of temperature and rock pressure, this subfacies probably represented higher water pressures than the green schist facies (Miyashiro and Seki, 1958).

When we consider the magnitude of rock pressure of this subfacies, the occurrence of jadeite is very important. From experimental and thermodynamic approaches, the stable association of jadeite-quartz and jadeite-quartz-sodic plagioclase require very high solid pressures which increase with rising temperature. In many jadeite-bearing crystalline schists, especially pelitic and psammitic schists, of the Sanbagawa belt, jadeite is stably associated with quartz and sodic plagioclase.

The thermodynamic and experimental studies of jadeite by Fyfe (Fyfe, Turner and Verhoogen, 1958), Robertson and others (1957) and Adams (1953) show that, if the temperatures in this subfacies were, say, almost 200°C, the minimum solid pressure of the present subfacies is between 6 and 12 kilobars. The solid pressures of the order of magnitude of 6-12 kilobars correspond to load pressures at a depth of 20-40 km when the average density of rocks is considered to be 2.8.

It may be rather difficult to imagine that the rocks of this subfacies in the Sanbagawa metamorphic belt were in such a depth, because they grade into unmetamorphosed sediments within the distance of 1-2 km. However, when we assume that a thick pile of cold, wet sediments was rapidly depressed into a deep horizon during the regional metamorphism, these values of temperatures and pressures (200°C-300°C and 6-12 kilobars) may not be impossible.

Significance of lawsonite-pumpellyite-epidote-glaucophane subfacies in Sanbagawa metamorphism.—In the Kantô Mountains and Sibukawa district, there are all gradations in mineral assemblages and degree of recrystallization from the unmetamorphosed Paleozoic formation to the highest-grade part of the crystalline schists. It is reasonable to think that the temperature of metamorphism increased progressively from the lowest-grade part to the highest-grade one. The highest-grade rocks of these districts are similar to those of the green schist facies. No biotite was formed in these two districts. No rocks belonging to the epidote-amphibolite facies and amphibolite facies were found. Therefore, the maximum temperatures in these two districts were probably lower than those of the epidote-amphibolite or amphibolite facies.

There appear to exist, at least, three possible courses for the variation of pressure conditions in the Sanbagawa metamorphic terrains:

(1) It is conceivable that the rock pressure during the metamorphism may have been higher in areas of higher temperatures. If the phase relation of albite

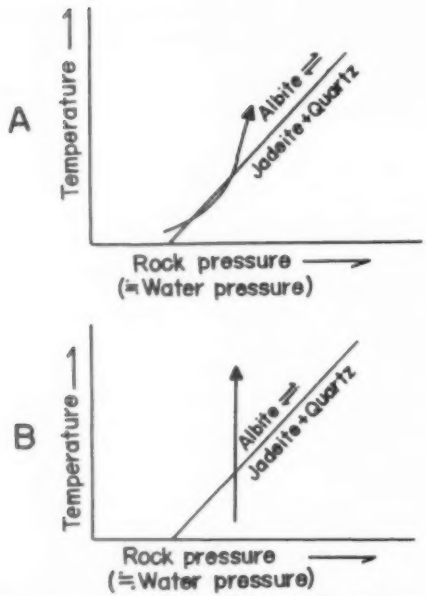

Fig 3. Probable temperature-pressure gradients in the Sanbagawa metamorphic belt inferred from the equilibrium slope of albite → jadeite + quartz.

A. Gradual increase of temperature, solid pressure and water pressure from unmetamorphosed part to the highest grade part.

B. Progressive increase of temperature under almost constant solid and water pressure.

and jadeite and the temperature-pressure gradient during the Sanbagawa metamorphism were as schematically shown in figure 3A, the occurrence of jadeite in the low-grade parts in the Sanbagawa belt would be well illustrated.

(2) Another possibility is a gradual rise of temperature under almost constant high rock pressure. In this case, we would expect the occurrence of jadeite not only in the lawsonite-pumpellyite-epidote-glaucophane subfacies, but also in the lower-grade parts (i.e., less-recrystallized and probably lower-temperature parts). Actually jadeite is absent in any rocks in such lower-grade parts. This may have been caused by a very slow rate of growth of jadeite under such low temperatures (probably $<200°C$) and weak differential movements.

(3) It is also conceivable that only the areas of the lawsonite-pumpellyite-epidote-glaucophane subfacies may have been prevailed upon by special high solid pressures and/or high water pressures. In this case, solid or water pressures may have decreased not only toward the unmetamorphosed sediments but also toward the higher-grade part of the Sanbagawa belt. In the Sibukawa district, many ultramafic rocks intruded into the areas of lawsonite-pumpellyite-epidote-glaucophane subfacies (Seki, Aiba and Kato, 1960, fig. 2). High solid or water pressures may have generated locally around these intrusive masses.

The present writer considers that we have a lot of things to do in the field and laboratory before any decision on this problem can be made.

ACKNOWLEDGMENTS

The present writer wishes to express his sincere thanks to the entire Petrological Laboratory in Geological Institute of the University of Tokyo for much kindness, valuable discussion and inspiration. The continued support and kind advice of Dr. Akiho Miyashiro and Dr. Fumiko Shidô must be particularly appreciated. The writer is deeply obliged to Mr. Mizuo Aiba and Miss Chigusa Kato, students of Saitama University, for their kind helpful cooperation in the field and in the laboratory.

A part of the expense of this study was defrayed by the grant for scientific researches from the Educational Ministry of Japan which is highly appreciated.

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[American Journal of Science, Vol. 258, December 1960, P. 716-727]

THE RELATIONSHIP BETWEEN PREFERRED ORIENTATION OF OLIVINE IN DUNITE AND THE TECTONIC ENVIRONMENT

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ABSTRACT. It is now well known that the olivine crystals of peridotites often have a strong tendency to a common orientation. Information on olivine fabrics was summarized by Turner (1942) with the addition of much new data, and Ladurner (1956) has recently surveyed the literature again and provided further examples of preferred orientation. Most of the studies of olivine orientation recorded in these papers have, however, been made without reference to the geographical orientation of the rock specimens in the field, so that the relationship of the axes of the oriented crystals to the boundary of the parent peridotite body, or to local tectonic directions, are not known (except in so far as fissility planes or chromite layers in the hand specimen are assumed to lie in particular tectonic orientation). The present contribution attempts to relate the orientation of olivine crystals in the type dunite, from Dun Mountain, Nelson, New Zealand, already made known by Turner (1942), to the tectonic setting of the Nelson peridotite belt.

GEOLOGICAL SETTING

The general geology of the region was described by Bell, Clarke, and Marshall (1911) for the New Zealand Geological Survey. Features of present interest are summarized below, and a modified version of the official map is given as figure 1.

Dun Mountain (3703 ft) is a hill, about 1¼ miles across, of olivine-rock, chromite-bearing in parts, lying within the complex belt of serpentinite and other rocks mapped as the "Mineral Belt" by Bell, Clarke, and Marshall. The "Mineral Belt" as a whole reaches a width of nearly 4 miles in the neighborhood of Dun Mountain, and strikes N55°E. Beyond the immediate area of Dun Mountain it extends some 50 miles NE to D'Urville Island in Cook Strait, and 30 miles SW where it is cut off by a major fault. Throughout its 80-mile length it is generally conformable with the strike of the enclosing sedimentary rocks (see Geological Map of New Zealand, 1:1,013,760, New Zealand Geological Survey, 1947).

The surrounding sediments were all grouped as the Maitai Series by Bell, Clarke, and Marshall (1911). Later work by the Geological Survey has, however, re-established an earlier subdivision into two groups, the Te Anau Series and the Maitai Series of Carboniferous-Permian age, as shown below (after Wellman, 1948, 1953):

Maitai group	7000 ft	Conglomerate, sandstone, and limestone.
	2000 "	Green and purple banded mudstone.
	7000 "	Gray banded mudstone and sandstone.
	2000 "	Limestone and calcareous sandstone (Maitai Limestone).

Te Anau group	5000 "	Pillow lava, conglomerate, and breccia.
	7000 "	Red and green sandstone, mudstone with plant impressions.
	20,000 "	Coarse sandstone, sandstone with fragments of black argillite, conglomerate, tuff and lava.

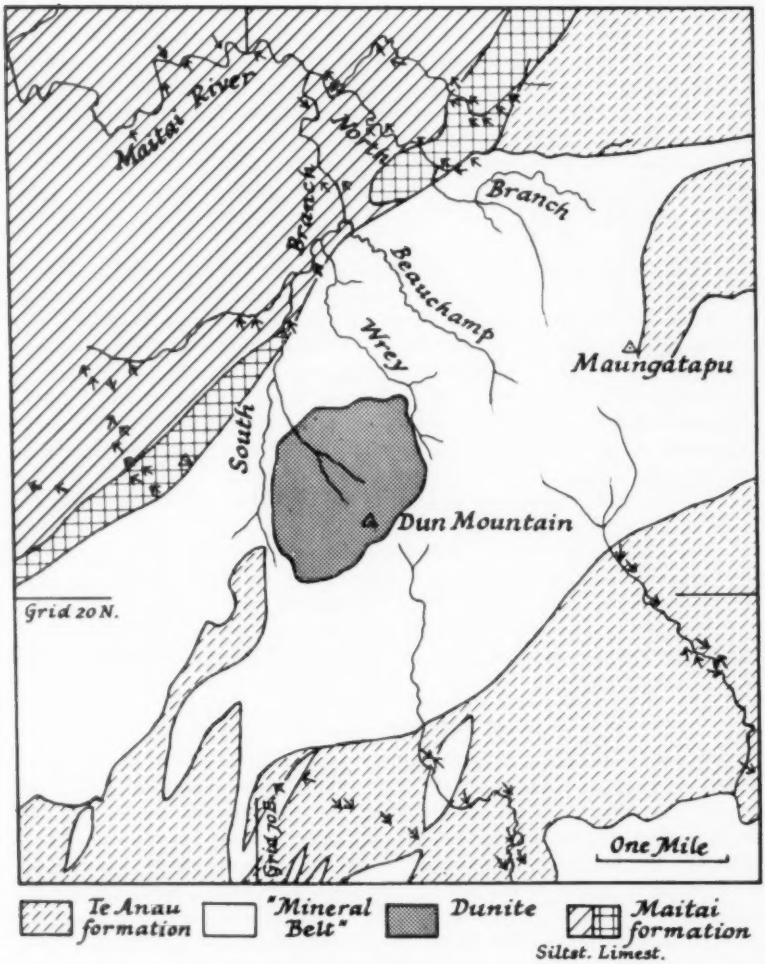

Fig. 1. Geological map of Dun Mountain area, Nelson, New Zealand, after New Zealand Geological Survey, with additions.

The rocks of the "Mineral Belt" are emplaced immediately below the horizon of the Maitai Limestone.

Passing south-eastwards, transverse to the regional strike, in the vicinity of Dun Mountain, the formations met with are:

(1) Siltstone, argillites, and sandstones of the middle part of the Maitai Series. These rocks are closely folded, individual minor anticlines and synclines being of the order of 50 to 100 feet across. Associated with this is a coarse fracture-cleavage parallel to the axial lines of the folds. Quartz "saddle-reefs" have sometimes segregated in the arches of the folds.

(2) A partly recrystallized limestone, the Maitai Limestone.

(3) Serpentinite, in generally concordant contact with the limestone, though showing much internal shearing and some involvement of limestone blocks. This junction is the boundary of the undifferentiated "Mineral Belt" of Bell, Clarke, and Marshall. Although these workers did not subdivide the "Mineral Belt", they recognized its heterogeneous character and the occurrence within it of many inclusions of Maitai rocks (as defined by them), now probably to be classed as of the Te Anau group. Their map indicates broadly this state of affairs.

Within this belt occur serpentinites in great variety, with cognate gabbros, saussuritized gabbros, rodingite, and veins of prehnite. Allocthonous inclusions comprise fine-grained flinty sediment—the so-called "baked argillite"—and reddish and greenish fine-grained lavas, sometimes with pillow structure, or in the form of what seems to have been glass-cemented breccia, or as masses of ordinary breccia. Blocks of these rock types, sometimes many yards across, are surrounded by serpentinite and appear to be isolated in it, forming a dismembered stratigraphy within the tectonically mobile serpentinite. All the inclusions seen belong to rocks of Te Anau type.

This complex extends to the crest of the Dun Mountain Range to the NE and SW of Dun Mountain itself.

(4) At Dun Mountain, however, the serpentinite gives place by a sharp junction or rapid transition to a core of unserpentinized dunite (cf. cross section by Finlayson, 1909, p. 356). The approximate outline of this dunite core is shown on figures 1 and 2. As far as is known, this dunite body does not enclose masses of country rock. Some 500 yards SE of Dun Mountain summit dunite gives place again to the serpentinite complex.

ORIENTATION OF OLIVINE

Oriented specimens were collected from the core dunite at the points shown on figure 2. Because of the distribution of good exposures, most of the samples come from toward the margin of the body, but the central part was sampled near the summit. Petrofabric diagrams, plotted in the lower hemisphere of a Schmidt net, and rotated so that the primitive circle is horizontal, were prepared for each specimen (figs. 3-6). Three diagrams are presented for each rock, based on the location of X, Y, and Z optical vibration-directions in 50 olivine grains, except in specimen 8207a where 100 grains were measured. As Turner (1942) points out, 50 grains is an adequate number to show the orientation pattern.

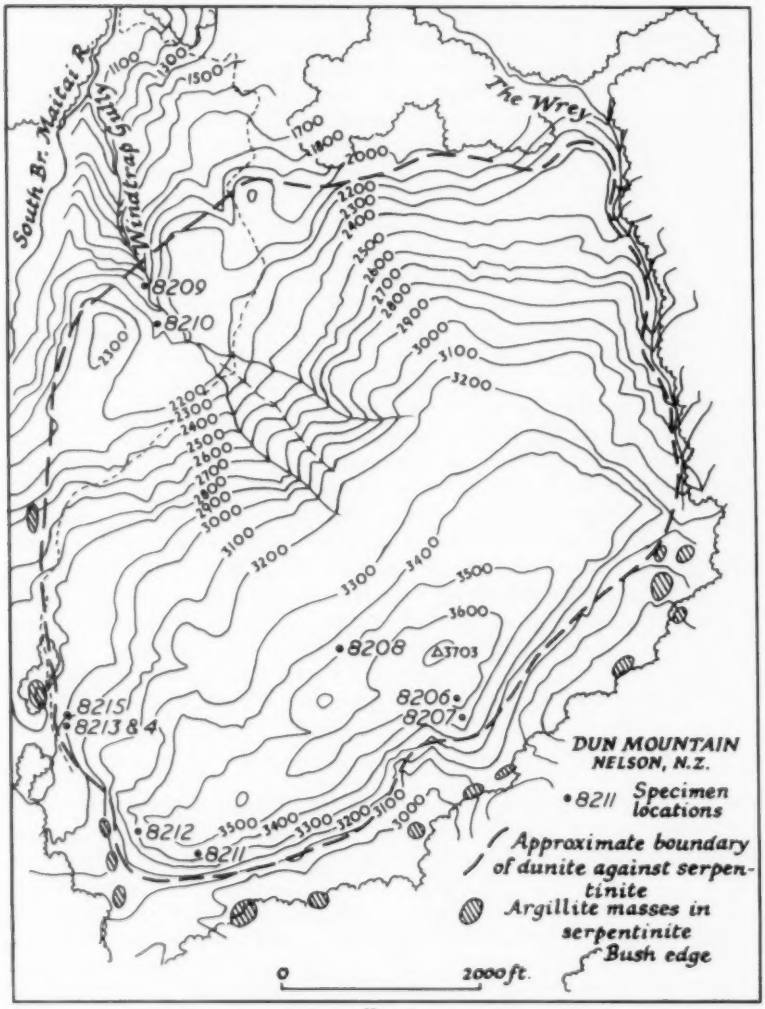

Fig. 2

The rocks have no visible schistosity, or fissility, and the grains are on the whole equidimensional. Dimensional orientation is recorded for olivine (Turner, 1942; Ladurner, 1956) and in particular for olivine in one specimen of dunite from Dun Mountain (Turner, 1942, p. 286); but in the rocks used for the present study sections cut at right angles to each other showed no obvious dimensional regularity, and the interlocking habit of the grains makes the value of statistical measurement doubtful.

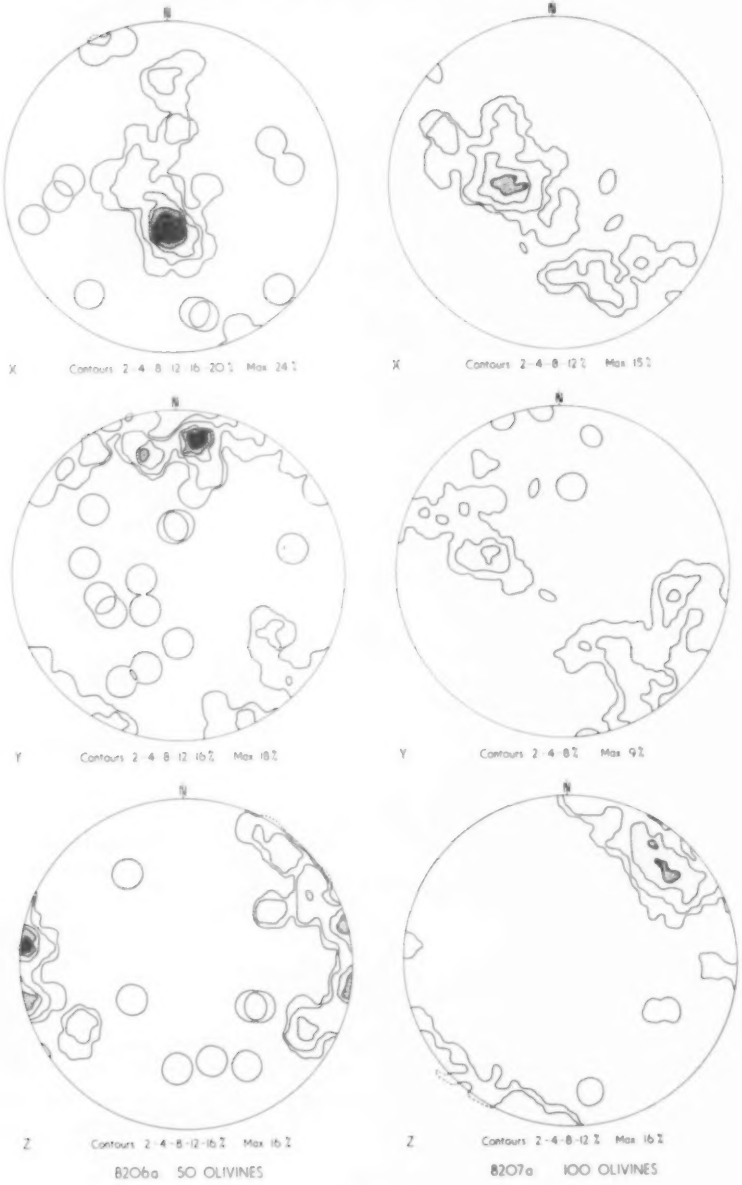
N
X Contours 2-4-8-12-16-20% Max 24%
X Contours 2-4-8-12% Max 15%
Y Contours 2-4-8-12-16% Max 18%
Y Contours 2-4-8% Max 9%
Z Contours 2-4-8-12-16% Max 16%
Z Contours 2-4-8-12% Max 16%
8206a 50 OLIVINES
8207a 100 OLIVINES

Fig. 3

The orientation diagrams all show a marked similarity, suggesting that the fabric is homogeneous throughout the mass. The symmetry of the pattern is orthorhombic. All the rocks examined have the X vibration-direction (= crystallographic *b*-axis) of the olivine concentrated in the central region of the stereogram, with maxima of from 12 to 24 percent per 1 percent area. There is a tendency for the spread of poles of individual diagrams to form a girdle toward NNW-SSE in the direction of the concentration of Y ellipsoid axis. Taken together, the X-maxima of all the diagrams lie in such a zone.

In every case, also, there is a concentration of poles of Z vibration-direction toward the margin of the stereogram. In five cases this concentration lies geographically NE-SW and more or less horizontal, or with a plunge which does not exceed 20°. Specimen 8206a (fig. 3) differs from the majority in having its Z-maxima E-W, while 8212b (fig. 5) has the maximum directed to S77°W, though a subsidiary concentration has the more usual direction. Maximum concentrations of Z range from 14 to 20 percent.

The concentration of Y is generally less than that of X and Z, except in the case of 8206 (fig. 3), a rock with a very strong preferred orientation altogether, where the Y maximum is as strong as the other two. Y tends to lie NNW-SSE with concentrations nearer the edge rather than the center of the stereogram. Except in 8207a and 8212b the horizontal disposition of the maxima is quite plain. The value of Y-maxima varies from 8 to 18 percent.

From these data it may be said that, in general, the olivine crystals of the Dun Mountain mass lie with optical Z (crystallographic *a*) directed between N40°E and N68°E. The average direction from five specimens, excluding 8206a and 8212b, is N55°E; from all specimens it is N66°E. X (=*b*) is more or less vertical, or steeply inclined, and Y (=*c*) roughly horizontal and directed NNW, though with some rolling of the crystal position about Z.

ORIENTATION OF JOINTS

The attitudes of the principal joints were measured at the locations of the oriented specimens and elsewhere in the dunite. Poles to 74 joints are plotted in figure 7, with great circles representing the planes to which the concentrations of poles correspond.

The concentration representing a plane striking N34°E and dipping 30°NW is the strongest. This may perhaps be correlated with the effect of the (010) cleavage of olivine normal to X vibration-direction in the crystal orientation diagrams. Thin sections show this parallelism of cleavage well when cut approximately normal to the statistical position of the (010) crystal plane.

The exact interpretation of the controlling factors in the formation of joints represented by the other four peripheral maxima is less certain. The maxima all lie in the NE quadrant and appear to form two pairs with average directions of N15°E and N85°E. However, two features which are certainly significant stand out. The pattern is symmetrically disposed about a line directed at N50°E and the symmetry, like that of the rock fabric, is orthorhombic.

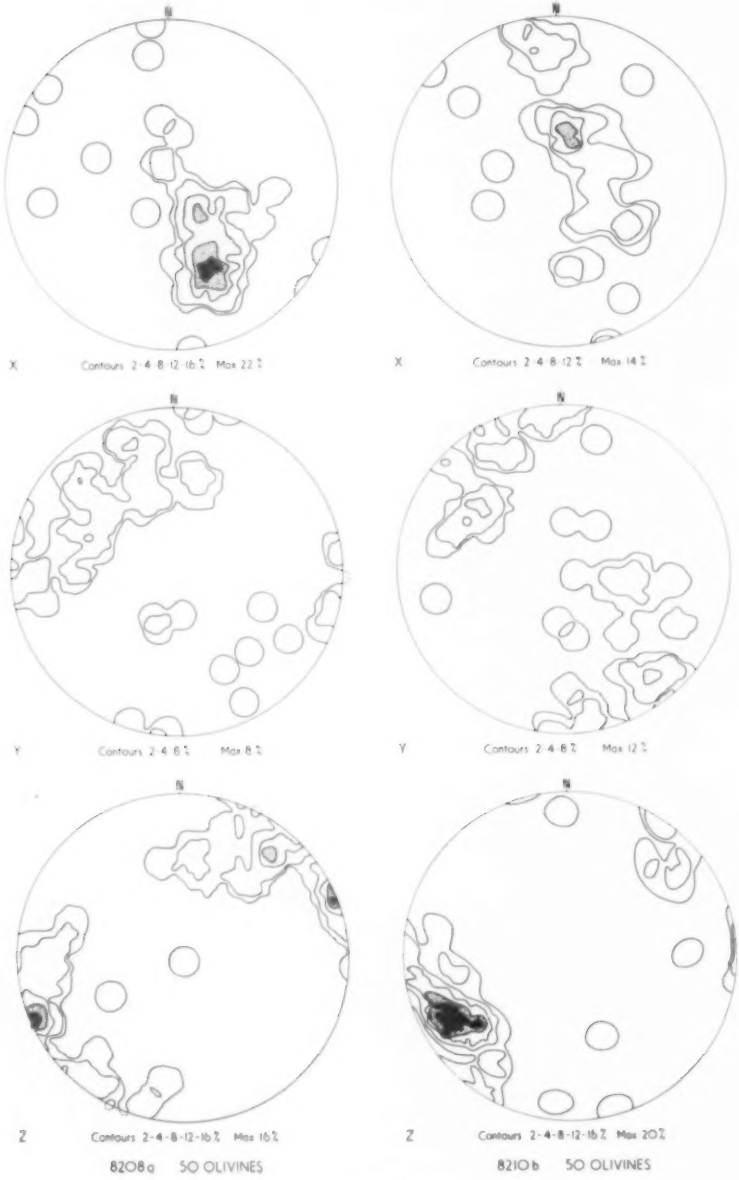
N
X Contours 2-4-8-12-16 % Max 22 %
X Contours 2-4-8-12 % Max 14 %
Y Contours 2-4-8 % Max 8 %
Y Contours 2-4-8 % Max 12 %
Z Contours 2-4-8-12-16 % Max 16 %
Z Contours 2-4-8-12-16 % Max 20 %
8208a 50 OLIVINES
8210b 50 OLIVINES

Fig. 4

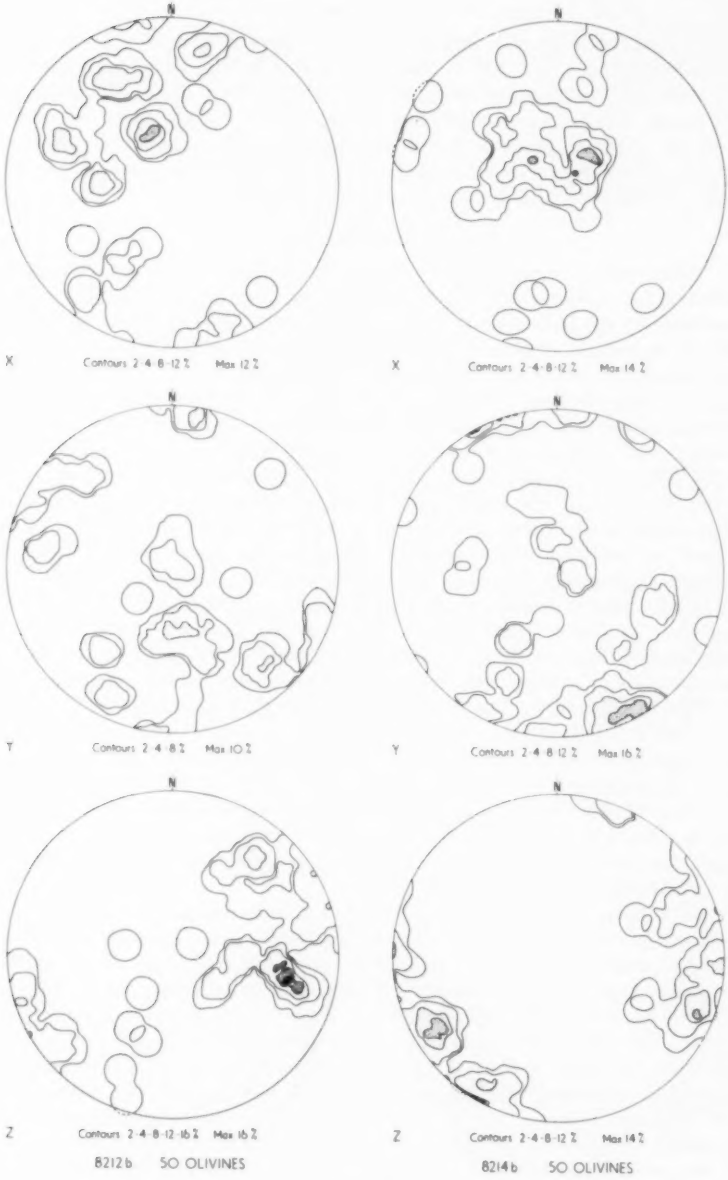
N
X
Contours 2·4·8·12 %
Max 12 %
N
X
Contours 2·4·8·12 %
Max 14 %
N
Y
Contours 2·4·8 %
Max 10 %
N
Y
Contours 2·4·8·12 %
Max 16 %
N
Z
Contours 2·4·8·12·16 %
Max 16 %
8212 b
50 OLIVINES
N
Z
Contours 2·4·8·12 %
Max 14 %
8214 b
50 OLIVINES

Fig. 5

FOLD-AXES IN ADJACENT SEDIMENTS

The mapping by the Geological Survey shows clearly the regional strike of major rock units at N55°E. Measurements of bedding planes and intersections between bedding and cleavage in the Maitai River (fig. 7) indicate that the sharp, upright, minor folds there plunge at 15° to N56°E in conformity with the regional trend of the major stratigraphic groups. R. N. Brothers (personal communication), using independent observations analyzed by the "beta-method", deduces an axial plunge of 12° to N50°E.

DISCUSSION

From the foregoing data it would seem that there is a definite relationship between the fold-axes, the olivine orientation, and the joint pattern of the peridotite in the Dun Mountain area. The correspondence in strike and in order of magnitude of plunge of the fold-axes and of optical Z of the olivines can hardly be fortuitous. The generally steep plunge of X in the olivines is also a striking feature. It would, of course, be of great interest to multiply the number of oriented specimens, both to confirm the apparent trends and especially to see whether the X-maxima of a larger sample align themselves along a NW-SE girdle, as seems possible from the present diagrams. Were this so, it would indicate that the (010) plane in the olivine was behaving like the bedding planes in the Maitai formation. A comparable tendency to girdling of the normal to (010) in individual rock specimens has been noted already (see Ladurner's summary, 1956, p. 34-35). Incidentally, in specimen 8206a (fig. 3), with a strong X-maxima (24%), undulose extinction bands (translation lamellae) parallel to (100) are strongly developed, as they are in all the rocks here studied. The correlation between strong development of (100) translation lamellae and a change from an X-maximum to an X-girdle mentioned by Phillips (1938, p. 134) does not seem to apply to the present case.

The two outstanding geological questions on which enlightenment might be sought from petrofabric study are the date and the mechanism of emplacement of the dunite. Bell, Clarke and Marshall (1911, p. 39) state that "there is in the 'Mineral Belt' rocks no sign of mineral change resulting from stress, nor is there any development of cataclastic or schistose structure." They conclude that the rocks are intrusive and were intruded later than the folding of the Maitai formation.

The present work shows that there is a definite relation between the fold-axes and the olivine orientation. Evidence of strain is found in the presence of undulose extinction lamellae and actual rupture of grains on (100) planes. This would be equally consistent with prekinematic or synkinematic emplacement of the dunite. While postkinematic emplacement along a privileged path parallel with the regional strike cannot be absolutely excluded, the orientation pattern is not easily explained on this assumption, and on general grounds of association of peridotites with orogeny it seems very much less probable.

Although Brown (1957, p. 16) is inclined to ascribe the orientation found by Phillips (1938) in peridotite from Rhum to sedimentary processes, to do so in the present case would be to ignore the (100) undulose extinction bands (also found in Rhum (Phillips, p. 133)), which are certainly due to strain.

X
Contours 2-4-8-12 %
Max 16 %
Y
Contours 2-4-8-12 %
Max 16 %
Z
Contours 2-4-8-12 %
Max 14 %
8215 a
50 OLIVINES

Fig. 6

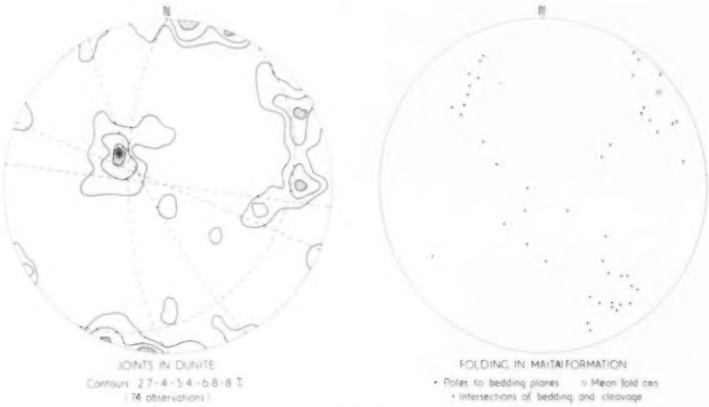

Fig. 7

At Dun Mountain the olivine orientation is undoubtedly due to pressure and most probably to the tectonic folding forces. The work so far done does not, however, permit the separation of any crystal-orientation effect due to forces of emplacement from that due to folding and thus does not serve to confirm or deny the possibility of quasisolid injection of dunite. It might have been expected that the envelope of serpentinite around the Dun Mountain core would, by its incompetence, have shielded the dunite from the effects of tectonic compression. It does not seem to have done so. Of course much, or all, of the serpentinization may have been later than the imposition of the orientation pattern. On the other hand, the penetration of the serpentinite into the Te Anau sedimentary and volcanic sequence, leading to the incorporation of so many inclusions of those rocks, suggests tectonic mobility of the serpentinite and thus its development before the end of the folding movements.

CONCLUSIONS

The discussion tends to the conclusion that olivine orientation at Dun Mountain is related to the axes of regional folding and is very probably produced by the folding forces. We are thus able provisionally to relate one commonly recorded scheme of olivine orientation to the local tectonic axes. It is found that optical Z (= crystallographic *c*-axis) of olivine may be oriented parallel to tectonic *b*-axis (the axis of folding) while X (= crystallographic *b*) may lie in the axial plane of upright folds, though it may also tend to form, with Y, a girdle in the *ac* tectonic plane. This represents an advance upon previous relating of crystal orientation to various types of s-planes.

Further petrofabric work on the Dun Mountain dunite, in conjunction with field studies on the dates of emplacement and, serpentinization would seem likely to clarify the mechanism of orientation further.

ACKNOWLEDGMENTS

The cost of the field work was met by a grant from the Hutton Fund of the Royal Society of New Zealand, to whose Council grateful acknowledgment is made. Thanks are also extended to Dr. R. N. Brothers of Auckland University for arranging for the use of a universal stage in the University Geology Department, and for his company in the field.

Much of the work was done while the writer was on the staff of Auckland War Memorial Museum, where the specimens studied are housed.

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[American Journal of Science, Vol. 258, December 1960, P. 728-739]

STRATIGRAPHIC RELATIONSHIPS OF THE LOWER ORDOVICIAN CHIPMAN FORMATION IN WEST-CENTRAL VERMONT*

W. M. CADY[1] and E-AN ZEN[2]

ABSTRACT. The Chipman formation includes four members: the Burchards, characterized by limestone with dolomitic mottling; the Weybridge, composed of limestone with thin interbeds of sandy limestone; the Beldens, made up of interbedded limestone and massive dolomite; and the Bridport dolomite member, consisting of massive dolomitic beds, which in the Cornwall area interfinger with limestones of the Chipman formation. Nearly a century ago Augustus Wing correlated strata of the Weybridge and Beldens with those of the Bridport dolomite. This correlation has been recently reaffirmed by Marshall Kay, who also recognized the faunal affinity of the Weybridge member of the Chipman with late Beekmantown age. Recent mapping by the authors, particularly by Zen in the Sudbury-Brandon area, shows that the sequence of the Burchards, Weybridge, and Beldens members, though generally in about that order, is locally much varied; also the Weybridge is locally missing in western areas and the Burchards is missing in some areas to the east. The irregular distribution of the Burchards, characterized by localized fossils, suggests biostromal deposits. These deposits apparently formed offshore from the Bridport dolomite member, which was deposited in near-shore shallows that fringed the eastern Adirondack area.

INTRODUCTION

The Chipman formation of west-central Vermont is a rock unit in the uppermost part of the Lower Ordovician, of Beekmantown age, between the underlying Bascom formation and the overlying rocks of Middle Ordovician (Chazy) age. Its structural setting is the Middlebury synclinorium and adjacent areas to the west (fig. 1). Four principal types of rocks characterize the Chipman formation: buff-to-brown, sharply defined, and laterally persistent beds of massive dolomite; white to blue-gray limestone; gray limestone with thin interbeds of sandy limestone; and blue-gray limestone with irregular spots of dolomite that give weathered surfaces a mottled appearance. These lithofacies are interbedded in the type section of the Chipman formation at the north foot of Chipman Hill, 1.5 miles north of the village of Middlebury. The Chipman formation was first referred to as a group (Kay and Cady, 1947), but it is here reduced to the rank of a formation. More recent studies (Kay, 1950), which are to be discussed below, have shown the synchronous facies relationship of the Bridport dolomite of Cady (1945, p. 545-546) with the other lithofacies. The "Beldens formation", the "Weybridge member of the Beldens" (Cady, 1945, p. 548, 550-552), the "Burchards limestone" (Kay and Cady, 1947), and the Bridport dolomite are here redefined as members of the Chipman formation. The Weybridge and Burchards have not been mapped north of the Middlebury synclinorium, whereas the Beldens and Bridport are considered formations in northwestern Vermont (Kay, 1958, p. 79-80) that correlate in time with all or part of the Chipman formation.

The Burchards member of the Chipman formation is predominantly a blue-gray limestone with dolomitic mottling. It is missing from portions of the east limb of the Middlebury synclinorium. The Weybridge member, which is

* Publication authorized by the Director, U. S. Geological Survey.

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characterized by gray limestone with thin interbeds of sandy limestone, is locally absent in the west limb of the synclinorium. The Beldens member consists of interbedded white to blue-gray marble and limestone and buff-to-brown massive dolomite that crop out throughout the belt of the Chipman. The Bridport dolomite member consists largely of massive dolomite and is the western facies of the formation.

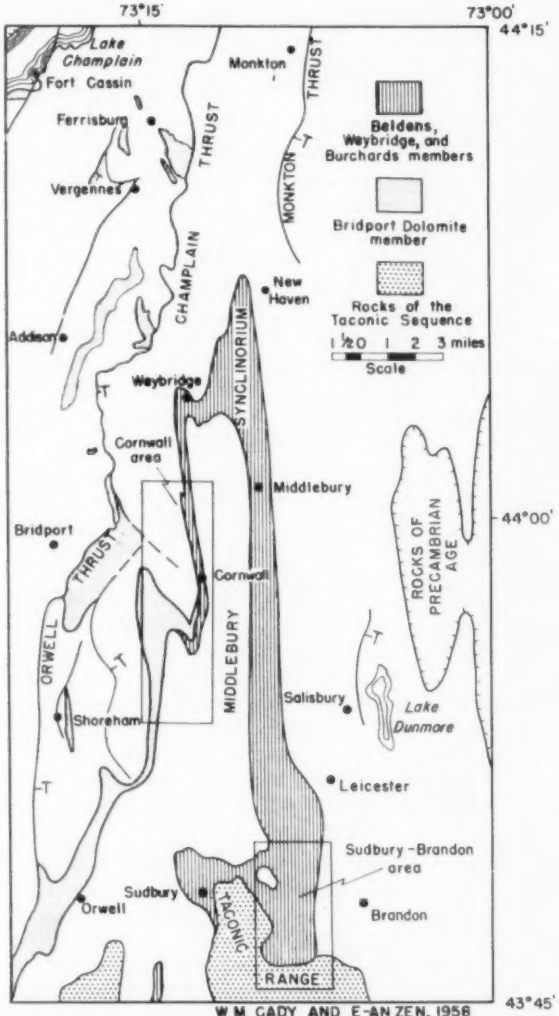

Fig. 1. Index map of the Middlebury synclinorium and vicinity, west-central Vermont, showing distribution of rocks of the Lower Ordovician Chipman formation.

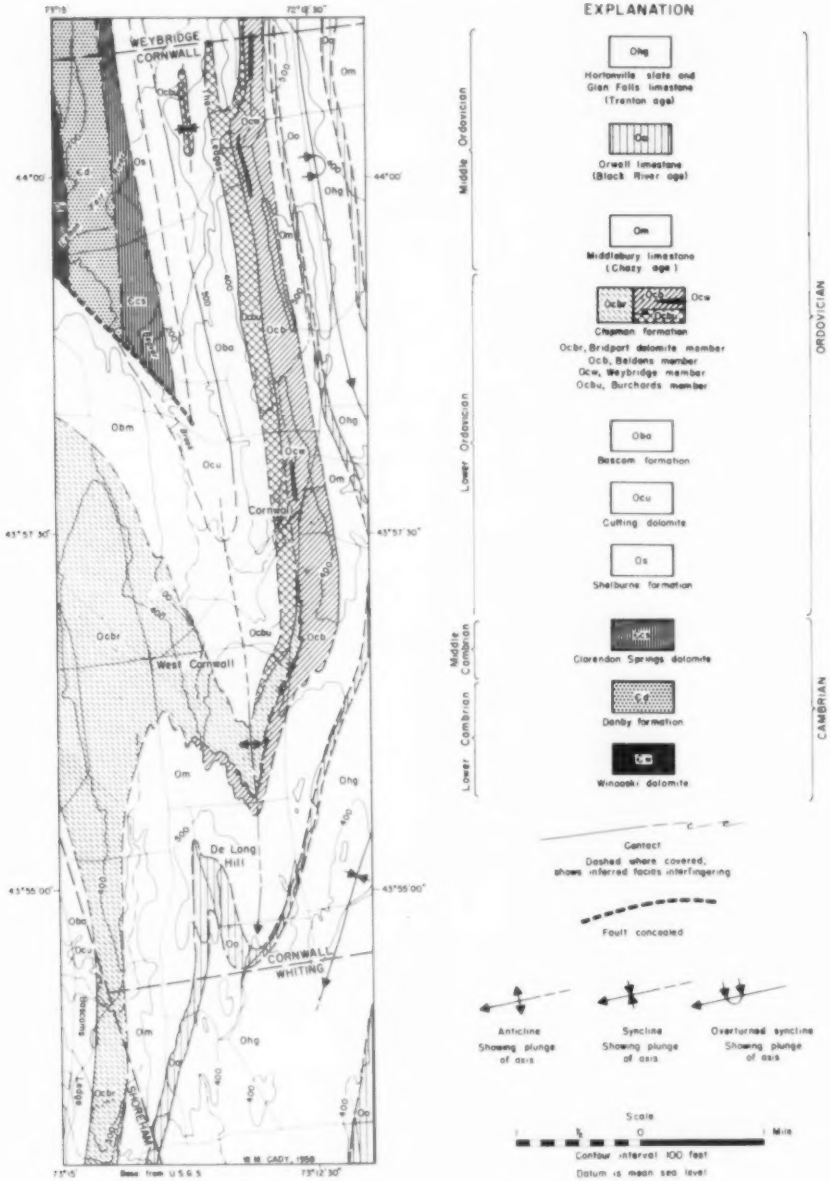

Fig. 2. Geologic map of the Cornwall area.

The Cornwall area, on the west limb of the Middlebury synclinorium (fig. 1), is most critical in showing the relationship of the Bridport dolomite member to the other members of the Chipman formation. Another critical area, which shows varied mutual relations of eastern lithofacies of the Chipman formation, is at the north foot of the Taconic Range between the villages of Sudbury and Brandon, on the east limb of the Middlebury synclinorium (fig. 1). The geologic maps (figs. 2 and 5) bring the mapping of these two critical areas up to date with the stratigraphic interpretations presented below, correcting earlier inferences (Cady, 1945, pl. 10) as to the areal distribution of the members of the Chipman formation, particularly of the Burchards and Beldens members.

CORNWALL AREA

The Beldens, Burchards, and Weybridge members of the Chipman formation are exposed in the northeastern part of the Cornwall area, and the Bridport dolomite member crops out in the southwestern part (fig. 2). Covering the northern part of the Bridport member are extensive surficial deposits that have made it impossible to prove correlation of the Bridport with the other members by walking out the units. Stratigraphic correlation is satisfactorily shown, however, by comparison of the sequences of formations in which the several members of the Chipman are contained. All units of the Chipman formation overlie the Bascom formation and underlie the Middlebury limestone.

The Chipman formation is exposed in a belt of discontinuous outcrops of rock of Beekmantown age that extends from about a mile south of the village of Cornwall north through The Ledges and into the town of Weybridge. The Chipman is in a section that, at the latitude of The Ledges, includes three Cambrian formations—the Winooski dolomite, the Danby formation, and the Clarendon Springs dolomite—as well as Ordovician formations of Beekmantown, Chazy, Black River, and Trenton age. The distribution and relationships of the stratigraphic units in this section are about as described and mapped by Cady (1945, p. 532-545, pl. 10), though their terminology is partly changed. The Shelburne is now referred to as a formation rather than a marble unit because it contains dolomite beds in addition to calcite marble, and also because limestone instead of marble occurs where the rock is unmetamorphosed, as in the Cornwall area. The "Crown Point limestone" becomes the Burchards member of the Chipman formation; and the Orwell limestone, like similar rocks that succeed those of Chazy age along Lake Champlain, is now assumed to be of Black River age rather than Trenton (Cady, 1945, p. 556; Kay, 1958, p. 91-92).

The Burchards member of the Chipman formation contains undetermined species of *Maclurites* and of a partly coiled cephalopod, *Eccyliomphalus*, such as occurs in upper strata of Beekmantown age. The *Maclurites* in particular are fairly abundant and can be seen in most outcrops. The Weybridge member, near the town line between Cornwall and Weybridge, contains the brachiopod genera *Diparelasma*, *Orthambonites*, and *Syntrophopsis* and the trilobites *Goniurus* and *Isoteloides*, forms which are diagnostic of rocks of late Beekmantown age (Kay, 1958, p. 79). The Beldens member, about a mile southeast of

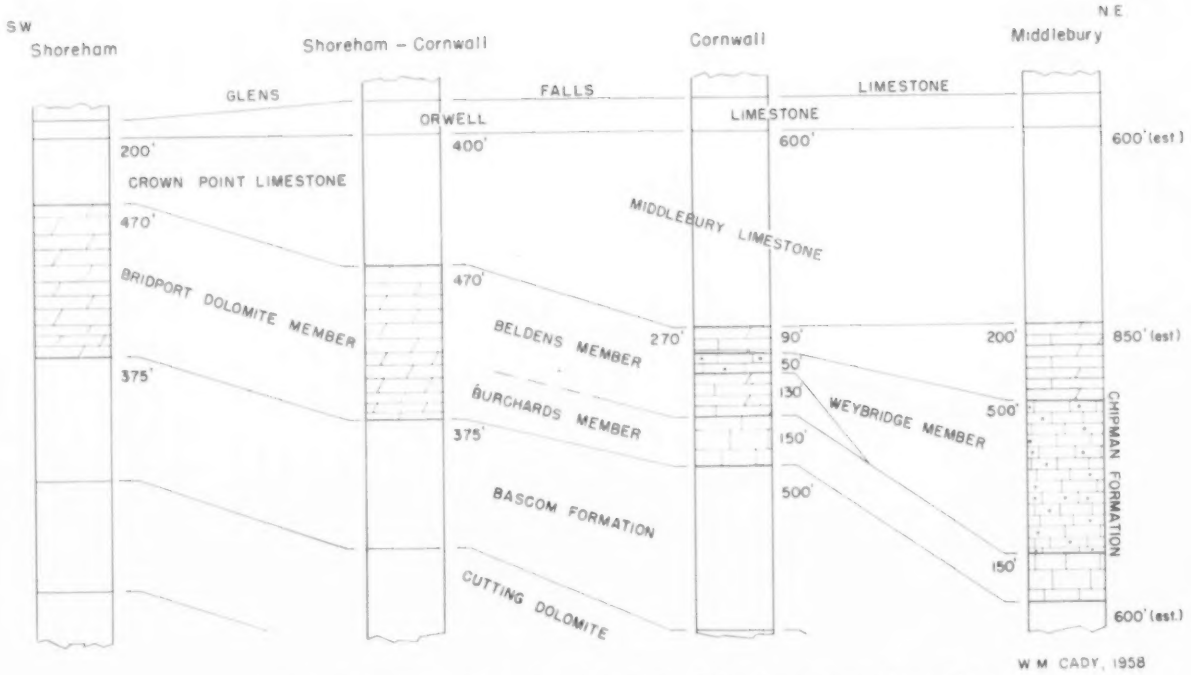

Fig. 3. Columnar sections showing stratigraphic relationships of the members of the Chipman formation and associated rocks between the villages of Shoreham and Middlebury.

Cornwall village, contains several undetermined orthoconic cephalopods but for the most part is barren of fossils.

The Bridport member of the Chipman formation appears at the extreme south end of the rock belt of Beekmantown age that passes through Cornwall village. A tongue of dolomite, which lacks interbeds of limestone such as characterize the Beldens, extends northward between the Burchards and Beldens members of the Chipman formation but pinches out about three-fourths of a mile south of the village. This relationship indicates that the Bridport interfingers laterally with the Beldens and Burchards members. This type of transition is inferred in an extensive area that is covered by surficial deposits, in the directions of De Long Hill and West Cornwall.

The principal exposures of the Bridport member in the Cornwall area are at Bascoms Ledge, which is in the vicinity of the eastern boundary of the town of Shoreham. The Bridport at Bascoms Ledge is in a section above the Cutting dolomite and Bascom formation (Cady, 1945, p. 541-545) of Beekmantown age and beneath the Middlebury limestone (Cady, 1945, p. 552-554), which is of Chazy age. The distribution and terminology of the stratigraphic units in the section near Bascoms Ledge, as shown in figure 2, are in part a revision of the former interpretation of Cady (1945, pl. 10).

The Cutting and Bascom formations and the Bridport dolomite member are as previously mapped (Cady, 1945) at Bascoms Ledge, but the Middlebury limestone is expanded westward and downward stratigraphically to the top of the Bridport and includes rocks that had been interpreted as "Crown Point limestone" and "Beldens formation". The Orwell limestone on the summit of De Long Hill, northeast of Bascoms Ledge, was misidentified as the Beldens during previous mapping. Also, the contacts between units in a covered area south of the summit of De Long Hill were incorrectly shown as solid rather than dashed lines. The inferred pattern of the Orwell limestone and of the overlying rocks of Trenton age (Glens Falls limestone and Hortonville slate) has been modified in this area.

The stratigraphic relationships of the members of the Chipman formation within, and also southwest and northeast of the Cornwall area, are shown in the columnar sections (fig. 3). The Cornwall section is a little north of the village of Cornwall and the Shoreham-Cornwall section is in the vicinity of Bascoms Ledge. The Shoreham and Middlebury sections are near the villages of Shoreham and Middlebury (fig. 1). Sections south of the Cornwall area on the west limb of the Middlebury synclinorium are like that in the vicinity of Bascoms Ledge.

The first, and apparently the correct, interpretation of the stratigraphic relationships of the Beldens and Burchards members to the Bridport member was arrived at nearly a century ago by Augustus Wing (Dana, 1877, p. 341-345), in the Cornwall area. One of the purposes of the present paper is to call attention to Wing's conclusions in further support of the inclusion of the Bridport as a member in the Chipman formation and of the time-stratigraphic unity of the various lithofacies of the Chipman. Figure 4 shows the correspondence between Wing's stratigraphic terminology and that used here.

		BASCOMS LEDGE AND VICINITY			CORNWALL VILLAGE TO THE LEDGES			
		CADY, 1945, 1960		WING, 1877	CADY, 1945, 1960		WING, 1877	BRAINERD, 1891
MIDDLE ORDOVICIAN	TRENTON	Hortonville slate		8 Hudson River slates	Hortonville slate		8 Hudson River slates	
	BLACK RIVER	Glens Falls limestone		7 Sparry limestone	Glens Falls limestone		7 Sparry limestone	[Not discussed]
		Orwell limestone			Orwell limestone			
	CHAZY	Middlebury limestone			Middlebury limestone			
LOWER ORDOVICIAN	BEEKMANTOWN	Bridport dolomite member of the Chipman formation		6. *Rhynchonella* beds	Beldens member / Weybridge mem. (Chipman formation)		6. *Rhynchonella* beds	"Chazy C"
					Burchards mem (Chipman formation)		5c. Coarse limestone	"Chazy B"
		Bascom formation	Zone 4	5b. Conglomerate	Bascom formation	Zone 4	5b. Conglomerate	"Chazy A"
			Zone 3	5a. Trilobite beds		Zone 3	5a. Trilobite beds	
			Zone 2	4. *Ophileta* beds		Zone 2	4. *Ophileta* beds	
			Zone 1			Zone 1		
		Cutting dolomite			Cutting dolomite			[Not discussed]
		Shelburne formation		3. Subcrystalline limestone	Shelburne formation		[Not discussed]	
CAMBRIAN		Clarendon Springs dolomite		2. Upper Potsdam	Clarendon Springs dolomite			
		Danby formation		1. Potsdam	Danby formation			
		[Not exposed]			Winooski dolomite			

Fig. 4. Stratigraphic terminology.

Wing (Dana, 1877, p. 341-342, 343-345) referred to both the Bridport and the Beldens as unit 6, the "*Rhynchonella* beds", and left no doubt that the two are to be correlated. Beneath the Beldens in the section near and north of Cornwall Village is the Burchards, a limestone that Wing referred to as unit 5c (Dana, 1877, p. 344). He mentioned the local occurrence of this unit in The Ledges north of Cornwall. Recent mapping shows that the Burchards is a lithofacies that is extensive enough to be mapped as a member of the Chipman formation north and south of the village of Cornwall. Also in the section near and north of Cornwall village is the Weybridge member (Cady, 1945, p. 550-551), which was referred to by Wing (Dana, 1877, p. 344) as the "striped stratum" in the "*Rhynchonella* beds". Wing also mentioned the "striped stratum" in the "*Rhynchonella* beds" at Bascoms Ledge (p. 343). There the "*Rhynchonella* beds" are now known as the Bridport dolomite member of the Chipman formation.

Brainerd (1891, p. 299) assigned a Chazy age to Zone 4 of the Bascom formation ("Calciferous D4") and to the whole sequence now known as the Burchards and Beldens members of the Chipman formation, in The Ledges north of Cornwall village. This incorrectly made these units younger than Bascom Zone 4 and Bridport (Beekmantown) of the section at Bascoms Ledge. The assignment of a Chazy age was predicated chiefly on the occurrence of coiled gastropods and cephalopods in the Burchards that in their poor state of

preservation resemble the gastropod *Maclurites magnus* of the type strata of middle Chazy age. Cady (1945, p. 548) recognized Zone 4 of the Bascom in The Ledges in Cornwall but followed Brainerd in correlating Wing's unit 5c (the Burchards) with the Crown Point limestone of middle Chazy age, and Wing's "*Rhynchonella* beds" (the Beldens) with strata of late Chazy age. The Beldens was mistakenly extrapolated into the section east of and stratigraphically above the Bridport southwest of De Long Hill (Cady, 1945, pl. 10). The rock above the Bridport here is actually the Middlebury limestone, which is of Chazy age but contains a few dolomitic zones that had suggested the "Beldens formation".

Kay and Cady (1947) "proposed that the 'Crown Point' formation east of the Champlain thrust in west-central Vermont be designated the Burchards limestone, named from a stream that enters the Lemon Fair River three miles west of the type locality in the belt of outcrop between Cornwall village and The Ledges." This seemed advisable because of the structural separation from the typical Crown Point limestone along Lake Champlain. Later Kay (1950; 1958, p. 79) recognized fauna in the Weybridge supporting late Beekmantown age, as well as the stratigraphic correlation of the Weybridge and the Beldens with the Bridport. Rock of Chazy age in the vicinity of Shoreham village is structurally isolated from the Middlebury limestone and is lithically and faunally like the typical Crown Point limestone; it is here continued as the Crown Point (fig. 3).

SUDBURY-BRANDON AREA

The geologic map of the Sudbury-Brandon area (fig. 5) includes most of the stratigraphic units that are in the section near and north of Cornwall village, as well as various rocks of the Taconic Range, which are shown as a single unit. The map shows several changes from previous mapping (Cady, 1945, pl. 10), notably discontinuous distribution of the Burchards ("Crown Point") member of the Chipman formation. The Chipman formation is underlain by the Bascom formation and overlain by Middle Ordovician limestone in the towns of Sudbury and Brandon much as it is in Cornwall. The rocks of the Taconic Range appear to overlie the Bascom and Chipman formations; their relationships are complex and are the subject of a continuing study by Zen.

The Bascom formation crops out three-fourths of a mile south-southwest of Arnold School and west of a secondary road, near the northeastern corner of the Sudbury-Brandon area. There the beds dip west in the east limb of the Middlebury synclinorium, and drag-fold relationships indicate that the tops of beds are to the west. The Weybridge member of the Chipman formation crops out about 500 feet west of the Bascom; 500 feet farther west near the Rutland Railroad, and at the eastern edge of the Brandon Swamp the Beldens member crops out. The Bascom and Chipman reappear, from beneath Long Swamp, at Halls Island one and one-half miles to the south. The unit directly above the Bascom at Halls Island is the Beldens member of the Chipman instead of the Weybridge member. Some of the limestone beds in the Beldens show dolomitic mottling such as is characteristic of the Burchards member.

Seager Hill, which is one and one-half miles south of Halls Island, is underlain by black slates that form part of the Taconic Range in the southern

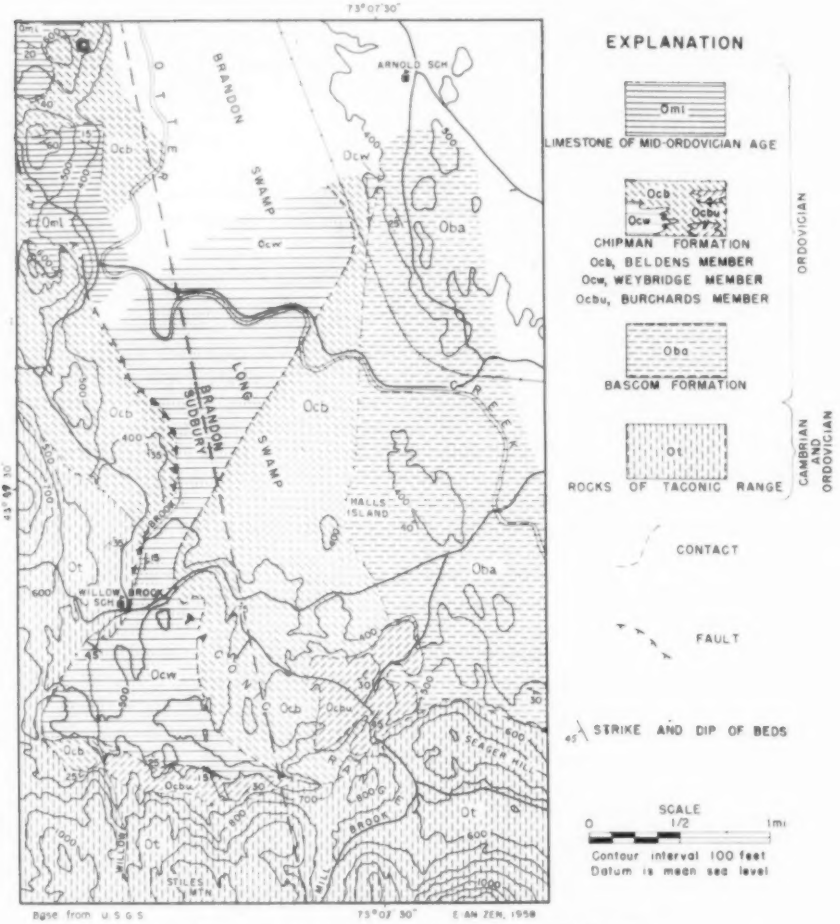

Fig. 5. Geologic map of the Sudbury-Brandon area.

part of the Sudbury-Brandon area. Both the Bascom formation and the Chipman formation are underneath the slate at the northwest side of the hill; the Bascom is east of the Chipman in apparent continuation of the trend southwest of Arnold School and at Halls Island. The Chipman immediately northwest of Seager Hill includes a large area of the Burchards member, which is characteristically mottled with dolomite. The Burchards of this area grades southward and westward into interbedded limestone and dolomite typical of the Beldens member. The Beldens, in turn, grades westward into the Weybridge member, which apparently overlies the Beldens conformably in the neighborhood of Willow Brook School.

A little south of Willow Brook School the Weybridge member strikes northwest and dips southwest, but north of Stiles Mountain it swings to an east strike and a gentle south dip and conforms to the trend of the contact between the Burchards member and the rocks of the Taconic Range. Crossbedding indicates that the section is right side up. To the south of the Weybridge, and apparently stratigraphically above it, are rocks of another area of the Beldens as well as of the east-trending belt of the Burchards.

The areal distribution and structural and stratigraphic relations of the Beldens, Burchards, and Weybridge (outlined above) indicate that these members are lithofacies in the Chipman formation. The sequence of these units varies; southwest of Arnold School the Weybridge member is at or near the bottom of the formation; at Halls Island and northwest of Seager Hill either the Beldens or the Burchards is at the bottom, and they are succeeded to the west and upward stratigraphically by the Weybridge; north of Stiles Mountain the Weybridge is succeeded by both Burchards and Beldens lithofacies. The time-stratigraphic position of the latter two is above that of the Beldens and Burchards at Halls Island and northwest of Seager Hill. The gradational contacts between units make it improbable that such repetition is produced by faulting. A schematic restoration of the Chipman formation, which interprets these relationships in a north-oriented section, is given in figure 6.

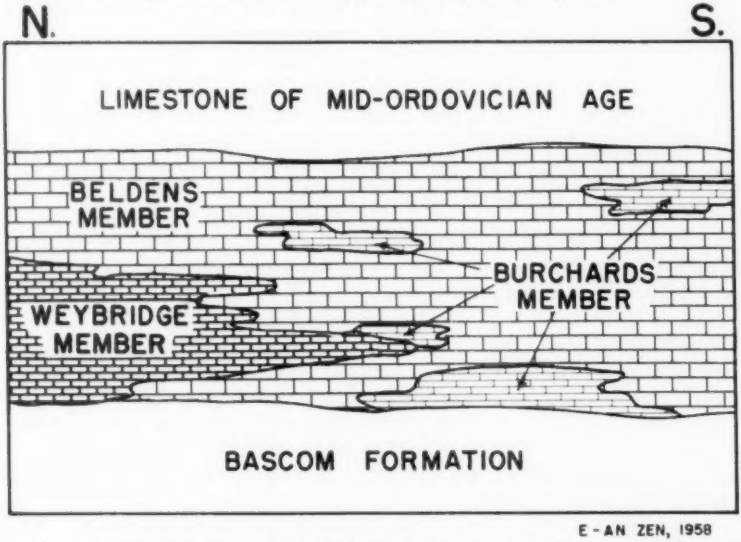

Fig. 6. Restored section of the Chipman formation approximately along the town line between Sudbury and Brandon.

The lithofacies of the Burchards member differs from that of the Beldens only in the numerous dolomitic spots that give the rock a mottled appearance. Close inspection of these spots reveals that they are remains of fossils. The spots are commonly so crowded together that they are actually in contact. It is

suggested here that the difference between the Burchards and the Beldens is primarily ecologic; the irregular distribution of the Burchards in the Sudbury-Brandon area suggests biostromes.

Small cystoid or crinoid stems have been found in the Weybridge and Beldens members of the Chipman formation, and sections of fossil shells are shown in the Burchards member by dolomitic mottling. The mineralogy of the Burchards limestone, particularly its relation to fossil preservation, is discussed by Zen (1959). Though specific identifications of fossils that might be useful in correlation have not been attempted, the lithofacies of the various units are so distinctive that correlation with the section near Cornwall is clear.

DISTRIBUTION OF FACIES AND PALEOGEOGRAPHY

The Chipman formation is a complex of limestone and dolomite facies; strata of massive dolomite, which are interbedded with limestone in the Beldens member, extend westward into similar dolomite that characterizes most of the Bridport member (fig. 3). The massive dolomite, as already indicated, forms sharply defined and laterally persistent buff-weathered beds as opposed to the mottled and irregularly distributed dolomite produced by alteration of fossil shells in otherwise nondolomitic limestone. Such westward increase in proportion of dolomite relative to limestone is generally characteristic of rocks of Beekmantown age not only in western Vermont but in other areas of the Appalachian region (Cady, 1945, p. 540).

The Burchards member of the Chipman formation, which shows the dolomitic mottling, is irregularly distributed east of the zone of transition into the Bridport member. The Burchards is predominantly beneath the Beldens member in the section near and north of Cornwall village, but in the Sudbury-Brandon area some of it, as already pointed out, also occurs above the Beldens. The Burchards appears to be continuous laterally in the Cornwall section but not in the Sudbury-Brandon section. These relationships plus the fossil content suggest that the Burchards is an irregular biostromal zone that formed east of and offshore from the massive Bridport dolomite member, which was deposited simultaneously in near-shore shallows.

The Chipman formation thickens eastward (Cady, 1945, p. 551) from the postulated biostromes, and in this direction nondolomitic limestone becomes more abundant in both the Beldens and Weybridge members. The sandy limestone of the Weybridge member thickens from 0 at the meridian of Shoreham to a maximum of 500 feet at the eastern limits of its exposure in the east limb of the Middlebury synclinorium (Cady, 1945, p. 551); it grades westward into the Burchards and the lower part of the Beldens and in some places persists even into the Bridport member.

The Beldens and local facies like that of the Burchards are found in other areas to the north and south of west-central Vermont. The Beldens is mapped in northwestern Vermont and adjacent Quebec as a formation (Kay, 1958, p. 68, 72, 76), and the authors have recognized it as far south as Berlin, New York. The Bridport, on the other hand, seems restricted to west-central Vermont and to nearby parts of New York that border the eastern Adirondack Mountains.

ACKNOWLEDGMENTS

The authors are indebted to John Rodgers, who pointed out several errors in Cady's earlier mapping of the belt between Cornwall and Orwell, and to A. L. Albee, A. L. Bush, A. H. Chidester, and Marshall Kay for critical reading of the manuscript. Distribution of the Bridport dolomite that is shown west of the Champlain thrust in figure 1 is from a manuscript map by C. W. Welby.

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[American Journal of Science, Vol. 258, December 1960, P. 740-743]

A POST-HARBOR HILL-CHARLESTOWN MORAINE IN SOUTHEASTERN CONNECTICUT*

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ABSTRACT. A discontinuous, northeast-trending belt, at least 13 miles long, of linear accumulations of boulders and patches of hummocky till at which outwash deposits head, lies about 8 miles north of Long Island Sound near New London, Connecticut. This feature, here named the Ledyard moraine, marks a temporary halt during ice retreat from the moraines of Wisconsin age of Long Island and southern Rhode Island. It lies south of the postulated line of the Middletown readvance.

A hitherto unrecognized halt in ice retreat in southern New England is marked by a discontinuous belt of accumulations of boulders and patches of hummocky till that extends for about 13 miles northeasterly across the Montville and Uncasville 7½ minute quadrangles, Connecticut, about 8 miles north of Long Island Sound near New London (fig. 1). As so far traced, the belt extends from near Ledyard Center in the town of Ledyard southwest across the Thames River near Gales Ferry and south of Uncasville to the Powers Lake and Pataguanset Lake area in the town of East Lyme (fig. 2). The belt is best marked by linear accumulations of boulders elongated along the trend of the belt, and is marked also by alignment of deposits of hummocky till and heads of outwash gravel. The belt is crossed by younger glacial and postglacial stream deposits. It lies roughly at right angles to the direction of ice movement as indicated by striae, grooves, and crescentic marks in the bedrock surface.

Linear accumulations of boulders consist of a great jumble of blocks up to 20 feet in diameter, but mostly less than 10 feet in diameter, without interstitial fine material at the surface (figs. 3 and 4). The boulder accumulations, where best developed, are between 200 and 400 feet wide and are as long as one-half mile, although linear accumulations of boulders and belts of very bouldery till can be traced continuously for about a mile. The distribution and trend of the boulder accumulations is independent of topography. The boulders in the accumulations are mainly granite gneiss, the most abundant boulder-forming bedrock in the area. The boulder accumulations cross diverse kinds of bedrock, some of which have little tendency to form boulders. Boulders of rock types foreign to most of the Montville and Uncasville quadrangles, which amount to less than 5 percent of the total, have been transported at least three miles from the north. Boulder accumulations locally merge into hummocky till, but more commonly the boulders become dispersed and the belt merges into bouldery smooth-surfaced till and becomes unrecognizable. Locally a belt of boulders is partly covered by penecontemporaneous or later outwash deposits in a valley. Linear accumulations of boulders are shown schematically on figure 2 by closely spaced x's; unusual concentrations of boulders in till or outwash are shown by more widely spaced x's.

Deposits of till form hummocky ridges mostly between 500 and 1,000 feet wide, locally with closed depressions. The till has a loose texture, is bouldery in most places, and locally contains lenses and blocks of stratified material. Texture and composition of deposits of hummocky till along the belt are not identi-

* Publication authorized by Director, U. S. Geological Survey.

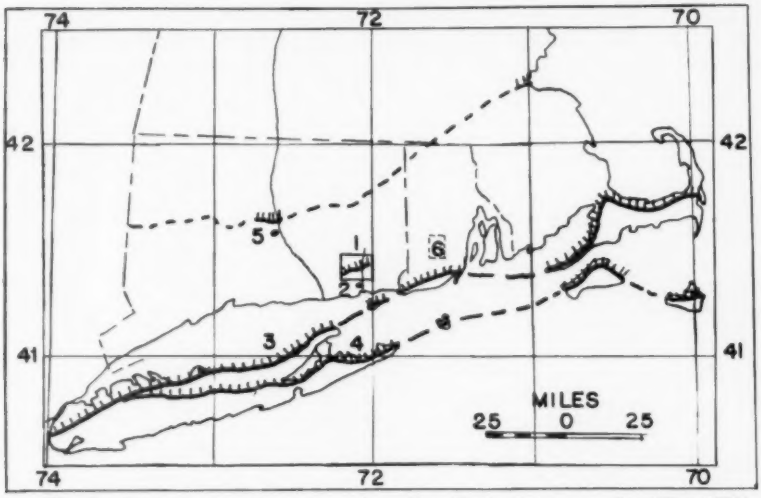

Fig. 1. Location of ice-fronts in southern New England (in part after Flint, 1953). 1. Ledyard moraine in Uncasville and Montville quadrangles, Connecticut, 2. New London, 3. Harbor Hill-Charlestown moraine, 4. Ronkonkoma moraine, 5. Middletown, 6. Slocum quadrangle, Rhode Island.

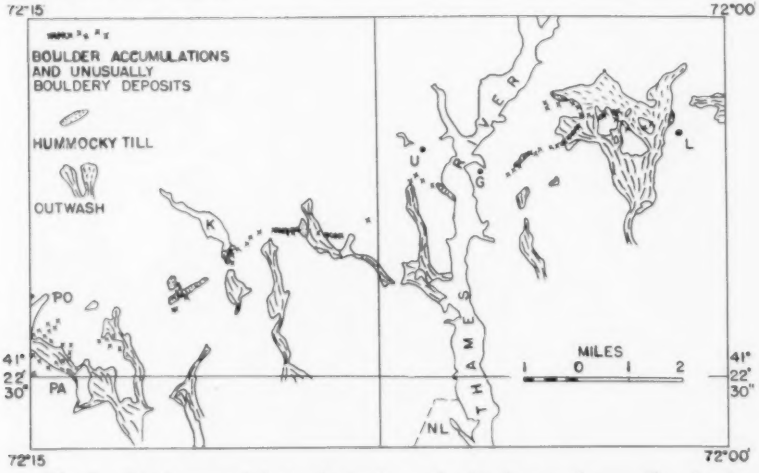

Fig. 2. Sketch map of Ledyard moraine and related outwash near New London, Connecticut. L—Ledyard Center, G—Gales Ferry, U—Uncasville, K—Lake Konomoc, PO—Powers Lake, PA—Pataguanset Lake, NL—New London. Only deposits heading near the Ledyard moraine are shown.

fied with certainty in every locality because of lack of internal exposures. The hummocky till may be ablation till, but its linearity suggests that it is an end-moraine deposit. Hummocky till is rare away from the belt.

The heads of several outwash deposits, some simple and others compound, lie along the belt. West of Ledyard Center, one of the largest outwash deposits has a compound head (fig. 2): one head, higher and older, is at the belt; the other, slightly lower and younger, is in a noteworthy kame-and-kettle area north of the belt. The two deposits merge southward, downvalley. The higher deposit overlaps the belt slightly but the overlapping part consists largely of sand and is interpreted as backfill of a ponded area left as the ice melted away from the belt. A smaller deposit with a compound head not indicated in figure 2 lies northeast of Pataguanset Lake.

The belt of boulder accumulations, deposits of hummocky till, and heads of outwash deposits are interpreted as marking an ice front. The boulder accumulations and hummocky till are end-moraine deposits. The feature formed by them is here named the Ledyard moraine after the town of Ledyard in which one of the boulder accumulations was first noted (Wells, 1890, p. 201). The moraine has been modified by stream action and probably is broken, in part, because of contemporaneous and later erosion. The lack of fine material in the boulder accumulations probably resulted from washing of till by meltwater during and after deposition. Lack of end-moraine deposits in some areas is possibly due to local instability of the ice front. The narrow width, intermittent character, and lack of widespread outwash deposits suggest that the moraine marks only a temporary stillstand, possibly following a slight readvance of the ice. No clear evidence of two ages of till has been observed in the area.

The Ledyard moraine is parallel to the larger well-known moraines of Wisconsin age of Long Island, southern Rhode Island, and Massachusetts (fig. 1), which, in contrast to the Ledyard moraine, consist mostly of outwash. The Ledyard moraine lies about 13 miles north of the Harbor Hill-Charlestown moraine, and about 20 miles south of the postulated line of the Middletown readvance (Flint, 1953, pl. 2; Ogden, 1959, p. 368), and appears to represent a temporary halt in the retreat of the ice northward from the Harbor Hill moraine. The Ledyard moraine when projected eastward falls within the Slocum quadrangle, Rhode Island (fig. 1), where Power (1957) mapped non-

Fig. 3. Boulder accumulation crossing valley northeast of Maynard Hill, 1 3/4 miles northeast of Gales Ferry, Ledyard, Connecticut. Larger boulders in foreground are about 5 feet in diameter. Swamp lies to left; outwash deposits to right beyond edge of photograph. Photo by G. L. Snyder.

Fig. 4. Detail of boulder accumulation in same locality as figure 3. Shovel on large boulder suggests scale. Photo by G. L. Snyder.

linear boulder accumulations and end moraine , some of which are part of the Queen's River moraine of Woodworth and Marbut (1896). These may be the equivalent of the Ledyard moraine. Intervening areas have not been mapped.

Recent pollen studies and radiocarbon dates of core samples from swamps in southern New England (Leopold, 1956; Deevey, 1958; Ogden, 1959; W. A. Niering, written communication) indicate oscillations of climate that probably reflect changes in regimen of ice during ice retreat and stagnation. From pollen analysis of a core taken from a swamp near New London, N. Beetham and W. A. Niering (W. A. Niering, written communication) recognize a climatic cooling which might relate to the formation of the Ledyard moraine.

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[American Journal of Science, Vol. 258, December 1960, P. 744-751]

REPEATED AVALANCHES AT CHAOS JUMBLES, LASSEN VOLCANIC NATIONAL PARK

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ABSTRACT. The Chaos Jumbles is an avalanche deposit covering more than two square miles. At least three separate avalanches descended at intervals of many years from the volcanic domes of Chaos Crags. All three flows are surprisingly similar as to extent and direction.

INTRODUCTION

The Chaos Jumbles is an avalanche deposit of dacite blocks and fragments strewn over more than two square miles in the northwestern part of Lassen Volcanic National Park. It has the typical hummocky topography of mudflows, but exhibits evidence of dry avalanche flow. A comprehensive study made by Williams (1928, 1932) showed that the deposit originated from the northern dome of the Chaos Crags as the result of explosions near the base. The resulting avalanche after moving downslope for about a mile swept upward for 400 feet on to the slopes of Table Mountain, more than two miles from the source.

Studies made by Heath (1959) indicated the age of the most recent avalanche as being roughly 270 years and suggested several older ones.

ANALYSIS OF AVALANCHES

The evidence which suggests that several avalanches occurred at widely different times is both qualitative and quantitative. The distinctly lighter portion of the aerial photographs (fig. 1)[1] can be readily noted in the central region of the deposits, and the traveler, having seen this, can distinguish it in the field though the difference is much more subtle than the photograph would suggest. Differences in the density of vegetation are not readily seen when the observer sights parallel with the terrain unless he makes a point of looking for them. Because of the subjective factor involved, an effort was made also to derive quantitative evidence to indicate the separate nature of the flows in an impartial manner. Each of these types of evidence will be considered in turn.

QUALITATIVE EVIDENCE

Examination of a great deal of the Jumbles area points to three separate avalanches that took place at quite different times. The most recent avalanche (fig. 1, boundary C) shows a paucity of trees and almost no shrubs at all. The dacite fragments are sharp and essentially unweathered and soil pockets of any kind are rare. This avalanche, which appears light in the photograph, produced a terminal mound roughly 120 feet high that served as a barrier to deflect the avalanche westward. Near the terminus, the avalanche deposit tends to blend with earlier ones but at least a mile of the north and east boundaries are traceable with great accuracy.

The middle avalanche deposit (fig. 1, boundary B) is characterized by taller and more numerous trees in some parts by a distinctive cover of bitterbrush (*Purshia tridentata*) and some other shrubs, and by dacite which is

[1] U. S. Forest Service CWX 11-143 and 22-119.

Fig. 1. Aerial photograph of Chaos Crags and Chaos Jumbles. The scale is the same as that for fig. 2. A is the edge of the most recent avalanche, B is the border of the middle avalanche and C is the edge of the oldest deposit near the area of maximum rise on Table Mountain.

definitely grayer and exhibits many weathered fragments. In many parts of this avalanche deposit, soil has developed in the interstices so that in the areas of heaviest cover there is an actual mat of needles and soil. The terminal ridge for the main course of this avalanche is not as high as that of the other two but it can be followed rather easily along the portions where it is highest. Early emigrants were sufficiently aware of this topography to run their road entirely to the north of this border, coming close at times but always remaining on the earliest avalanche deposit (see emigrant trail, fig. 2).

The first of the three avalanche deposits (fig. 1, boundary A) shows primarily on Table Mountain (fig. 2) and it was this one which extended nearly 400 feet up the slopes. Only in a few other areas is this deposit still well exposed although strips 50 to 100 yards wide appear around much of the periphery of the Jumbles. This deposit exhibits the greatest difference from the others; its cover is that of a nearly mature or sub-climax forest and most of the surface is mantled with soil. In addition, the hummocks on the surface are much more rounded and flatter than those elsewhere in the Jumbles, their slopes departing much farther from the angle of repose. Furthermore, scattered pieces of the Table Mountain lavas have drifted over the avalanche deposit in many places. Such transport much have required a long period of time.

Specific details on cover will be dealt with under quantitative evidence but certain peculiarities can best be noted here. Yellow pines (*Pinus ponderosa* var. *jeffreyi*) on the youngest avalanche deposit are usually dwarfted and distorted. On the second deposit, these trees are often normal in pattern but have sharply conical trunks that indicate exceedingly slow growth. On the oldest avalanche deposit, the yellow pines are magnificent specimens with no evidence of anything but the most normal growth. Another pine, the Western White (*P. monticola*), is also informative. Numerous specimens on the two youngest deposits are well developed, but the species is all but absent from the oldest deposit (table 1). It is typical of raw areas that they may be invaded by a variety of species so long as the primary competition is with the elements. Once soils have formed, competition becomes that of the biotic environment. These avalanche deposits demonstrate exactly this principle in operation.

TABLE 1

Average occurrence per 1000 feet of trees at least ten feet tall. Species: Pc, *Pinus contorta*; Pl, *P. lambertiana*; Pm, *monticola*; Pp, *P. ponderosa* var *jeffreyi*; Ac, *Abies concolor*; Am, *A. magnifica*; Ld, *libocedrus decurrens*.

Area	Newest	Middle	Oldest	Original
Based on	4000 feet	3000 feet	3000 feet	1000 feet
Pc	0.25	16.33	0.0	0.0
Pl	0.0	1.66	0.33	0.0
Pm	4.25	3.66	0.0	0.0
Pp	0.75	9.33	12.66	3.0
Ac	1.25	3.00	3.33	59.0
Am	0.75	2.0	0.0	0.0
Ld	0.0	0.0	0.33	0.0
Total	7.25	36.0	16.66	62.0

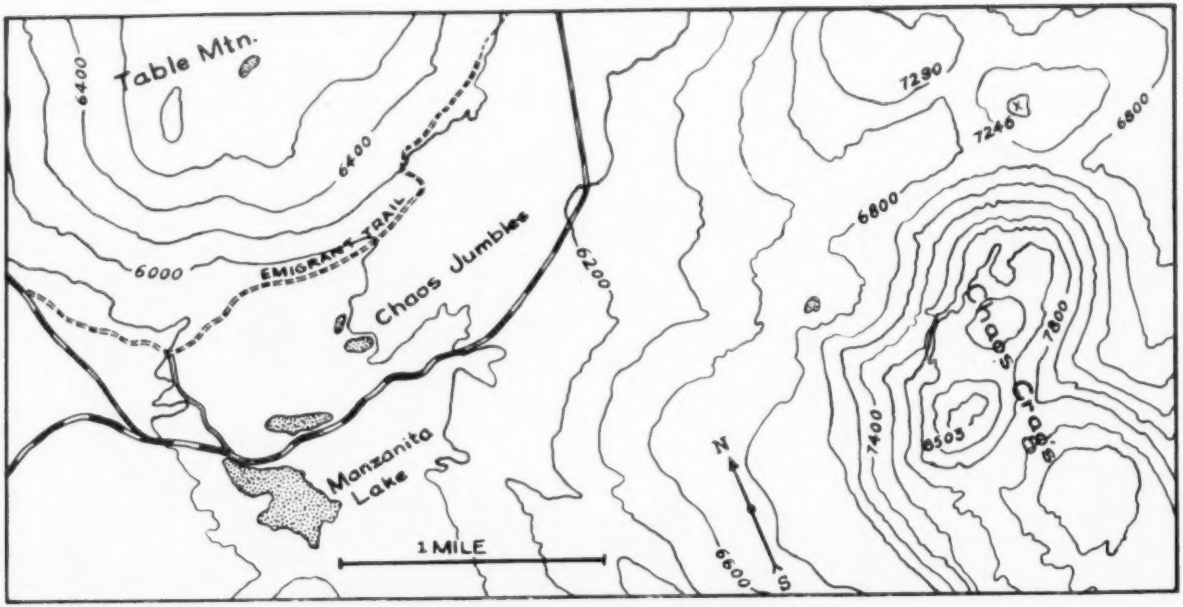

Fig. 2. A portion of the Manzanita Lake quadrangle (1956) which covers the same area as the photograph of fig. 1. Contours are at 200 foot intervals.

Another difference observed among these deposits is correlated with the severity of the environments. In the youngest deposit, there are many very small trees about six inches to four or five feet in height. These show a great deal of distortion and cores of some of the larger ones indicate that growth has been exceedingly slow. Some of these small trees and most of the larger ones demonstrate a gain of less than two inches of increment to the radius in a century. Cores from trees on the oldest avalanche deposit show very rapid growth (as much as two inches in less than ten years), the reduced rings appearing only as the trees gained maturity.

QUANTITATIVE EVIDENCE

The foregoing description of qualitative evidence has indicated some features readily noted by the field observer. However additional evidence of a quantitative nature is based on the effects of edaphic differences on cover. Plants are exceedingly sensitive to variations in soil quality and development. One of the standard methods of cover analysis is the line transect (Bauer, 1943) in which all plants which would touch a vertical plane along a particular line are recorded. The trees of the Jumbles indicate soil features and are relatively uninfluenced by short-term climatic fluctuations which could modify annuals or even perennial herbs with short life-histories.

A number of transects were run across the boundary of two deposits. Figure three displays to scale the considerable differences in the heights and numbers of trees on two of these 1000′ transects. Table 1 shows vast differences in tree cover on the different flows. Not only do the species differ, but the density varies markedly between flows. On the most recent flow, trees are few with the Western White Pine by far the most important species. The second deposit has many more trees with Lodgepole Pine dominant and with Jeffrey Pine a strong contender. On the oldest avalanche, the trees are very large and fewer in number. The Jeffrey Pine has the dominant position and the species dominants on the other flows do not even appear in the 3000′ of transects. A transect on the older soils adjacent to the avalanches shows strong dominance by White Fir. This suggests that even the oldest avalanche deposit is a very long way from being completely weathered to a mature soil.

Nearly one hundred cores were taken from the largest trees on the most recent avalanche deposit. These showed the oldest trees from an area of well over a square mile to be about 262 years old (Heath, 1959). Random sampling of but three of the trees on the oldest Jumbles indicated one tree to be 284 years old. Four trees close together on the second deposit all gave dates of more than 200 years and one was 261 years old. On the most recent avalanche, the dates show a rather sudden development of trees as could be expected on a relatively recent surface. Cores in the other areas where the oldest trees may have died and been destroyed cannot be used to date the time of origin of the deposits but they do serve to demonstrate their greater age.

OTHER FEATURES OF CHAOS JUMBLES

Williams' (1932) study was extensive and there is little point to reiteration of his report. However, the recognition of the separate flows does modify

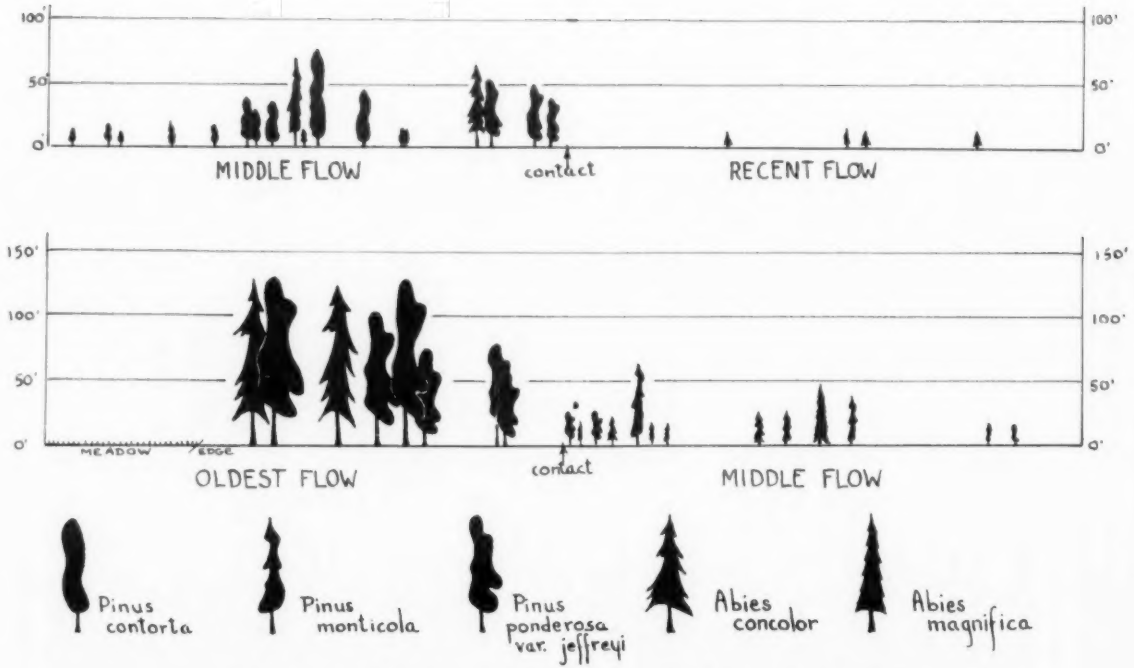

Fig. 3. Two 1000 foot transects from areas of contact between deposits. These are actual cases; not summaries.

some aspects of his interpretation. For one thing, the large moraine-like ridges so notable on the western border of the deposits are not the borders of the last flow but appear to be derived from the earliest one. The most recent flow built lower ridges parallel and nearby to these which accentuate the steepness and height of the old ridges.

Williams states,

> There is no marked increase in the size of the larger blocks as the flow is traced toward its source. All that can be said is that the sand-like fraction increases in amount toward the distal ends of the deposits.

This certainly applies well to the latest avalanche deposit. However, there are some larger rocks in the general area of the "crater" rims—perhaps due to being less subject to the battering received by rocks farther along. The "sand-like fraction" is a prominent feature of the region of the terminus and contributes greatly to the difficulty of determining this border which is so distinct in the mid and upper areas. The main linear flow built ridges some 120 feet high and these deflected the remaining rush to the westward. The increasingly smaller size of the material and the suggestion of braiding shown on the aerial photograph (fig. 1) along with the ill-defined terminal border imply a high moisture content and fluidity in these terminal portions. The gradient in these deflected materials is considerably less than that of the main linear flow and this also implies a well lubricated mass. These features tend to reinforce Williams postulation that a high water content was involved in the formation of the avalanche deposit.

Williams describes tumulus-like mounds which he postulates were derived from the crumbling of large blocks subjected to gases and steam before emplacement and subject to rapid disintegration after it. Three features would seem to deny this origin. Firstly, there is the paucity of large blocks in almost the entire deposit. Certainly a rock large enough to produce these mounds would be out of all proportion with the general mass of fragments composing the majority of the area. Secondly, various small areas of what might be termed "boil-ups" appear frequently in some parts of the newest Jumbles. These rather flat areas appear to be similar to mud mounds squeezed up in a bog. It can be assumed that some portions of the flow were wetter than others and in the churning mass were squeezed up in heaps. The small size of the materials leads to rapid leveling of the mounds so that lesser ones are almost flat. Thirdly, in all but one observed case, the nature of the materials, except for size, appears to be consistent with the rest of the flow. There is no excessive weathering of the fragments nor do these show particular evidence of steam or gas action.

A peculiar feature of the avalanches is the highly specific direction of flow of each. On three widely separated occasions, the dome must have risen enough to provide not only the huge mass of material but also the momentum required. Added to this is the requisite of some explosions on the north side of the dome such that the materials would topple in only one direction. There is no evidence of anything but very localized talus around the rest of the base of this northern dome so very violent explosions can be ruled out. Thus, we can accept the idea of the steam explosions Williams postulates but we must add

to this that they occurred not just once but three times. The "crateriform depressions" at the dome base would certainly appear to confirm this origin for the last flow. However, flow patterns of the avalanche materials can be traced to some extent by observing streaks of rock of slightly different color or grade (slightly visible in fig. 1). Some streaks are nearly half a mile long and consist of rock which appears to have been somewhat discolored by gases; others are merely streaks of consistently coarser or finer debris that can be traced over the highly irregular surface of the Jumbles, even into the inner rim of the "craters". Had the explosions thrown out such material, these flow streaks would not be present. Furthermore, there is little evidence on the surrounding tuffs, of dacite fragments blasted out onto the surface. In whatever manner the explosions occurred, the process must have stopped shortly before the avalanche ceased its flow and must never have been violent enough to do more than clear two pockets at the base of the dome. One is forced from all of this to postulate some rather improbable events. First, that the steam explosions each time were almost horizontal in their force since they did not cast debris onto the lateral tuffs. Second, on the last avalanche the explosions must have stopped just prior to the end of the movement since the craters are preserved yet the flow streaks extend into them. Third, we must postulate that similar blasts took place twice before in an almost identical manner.

A particularly interesting problem is posed by these separate avalanches with respect to the surrounding territory. On the west, as Williams notes, the deposits overlie tuffs from what appear to be rather recent explosions from a series of craters between Lassen Peak and Chaos Crags; craters which apparently extended over the older southern dome. These deposits must be a good deal older than was originally supposed.

There does not seem to be any present way to date the older flows. One can only note that soils form exceedingly slowly at these altitudes. Strictly from a biologist's estimate, it would appear that the second avalanche took place not less than 700 years ago and the oldest one not less than 1000 years earlier than that, but these are strictly subjective estimates.

ACKNOWLEDGMENTS

Dr. Gordon Macdonald was most helpful in the field in discussing the evidence. The National Park Service personnel and particularly Superintendent E. P. Freeland, Park Naturalists V. R. Bender and Richard Prasil and District Ranger Lester Bodine were most helpful in providing suggestions and encouragement, facilities and transport of equipment. Dr. Howel Williams read parts of the manuscript and offered many helpful suggestions.

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[American Journal of Science, Vol. 258, December 1960, P. 752-756]

MOVEMENT OF ROCKS BY UPROOTING OF FOREST TREES

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ABSTRACT. Boles of tall trees with large diameters behave as cantilever beams possessing great strength and the capacity for exerting tremendous leverage at the point of support or fulcrum. When trees are uprooted by wind they may move, both vertically and horizontally, rocks having a volume of as much as 50 cubic feet and a weight of around four and a quarter tons. Uprooting of trees contributes to downslope movement of the soil and rock mantle and locally may initiate erosion of the gully type. Some of the effects of soil and rock movement by uprooting trees are similar to those resulting from periglacial frost heaving.

INTRODUCTION

Uprooting of trees is a well-known occurrence in most forest regions and is especially common in places exposed to high winds. Trees growing on sites with shallow soils are more likely to be windthrown than trees in deep soils. Quite commonly topographic exposure to strong winds and shallow soils go together, as for example, ridges and upper slopes with lithosols. Of course trees may be uprooted on even the deepest of soils when wind velocities become unusually high.

Some of the effects of forest soil disturbance by uprooting of trees were described by Lutz and Griswold (1939) and Lutz (1940). During the course of that work it was noted that in some instances glacial boulders two or more feet in diameter were moved several feet vertically or horizontally when forest trees were uprooted. It appeared that when trees are windthrown a considerable amount of mechanical work may be accomplished through the rending of solid rock and movement of rock masses. Observations on the mechanical or geological work done when trees are uprooted were continued and the results are presented here.

REVIEW OF LITERATURE

Movement of rocks by uprooting of trees, although very common, has received only scanty attention in the literature. John Evelyn, in his *Silva* published in 1776, mentioned the loss of trees in a storm (p. 634)

> . . . several of which, torn up by their fall, raised mounds of earth near twenty feet high, with great stones intangled among the roots and rubbish . . . .

During his travels in North America, 1823-1827, David Douglas observed the mechanical work done by windthrown trees along the Columbia River. In his *Journal* one finds the following statement (p. 251):

> On both sides high hills with rugged rocks covered with dead trees, the roots of which being laid bare by the torrents are blown down by the wind, bringing with them blocks of granite attached to their roots in large masses, spreading destruction before them.

De Costa (1869) took note of the mounds of earth and rocks formed by uprooting of trees in the Lake George region in New York, recognizing them for what they were. That these features are sometimes mistaken for the work of man is indicated by De Costa's comment (p. 64):

> In the summer of last year (1867), the author saw a party of antiquarians opening one of these mounds. A large stone lay at each end, fully persuading them that the mound covered an Indian grave. Of course nothing was found.

PLATE 1

Masses of schist moved about 5 feet when a 15-inch chestnut oak tree and a 7-inch sugar maple were uprooted. The large perched block has a volume of 6.7 cu. ft and weighs 1130 lbs. Total volume of rock moved was 27.4 cu. ft, weight 4610 lbs.

PLATE 2

Partial uprooting of a 15-inch diameter scarlet oak tree raised a block of granite (9.0 cu. ft, 1520 lbs.) about 2 ft. The oak tree was hung up when its crown lodged in background trees.

Van Hise (1904, p. 446-447) in his classic treatise on metamorphism recognized the mechanical work performed when trees are uprooted, and wrote as follows:

> When a tree is overthrown by the wind, or in some other way, a large mass of the soil, subsoil, contained boulders, and even the solid rock, may be lifted 1, 3, or 5 meters above the surface level. When the trees decay the inorganic material again joins the soil, producing a mound. The material of the mound is in a much disintegrated and decomposed state; for it has been in a position to be effectively acted upon by all forces of weathering. How important this effect is can be appreciated only when one travels through the original forests. In such places there are seen almost everywhere hollows where trees have been uprooted and mounds where the material has fallen to the surface. Where tornadoes have swept through the forests, all the trees in their paths have been overthrown at once, and it seems as if almost the entire mass of soil and rock to the depth of several meters has been upturned.

A special case of rock movement involving uprooted trees is the long-distance rafting of boulders emmeshed in the roots of drift logs. It is probably a fairly common occurrence along streams where bank cutting is active, especially during floods. Walther (1893-1894, p. 648) recognized this mechanism of rock transport as also did Price (1932). The latter author observed erratic boulders, ranging in size up to 161 pounds, in the Sewell coal bed at Leslie, West Virginia. The nearest source of these boulders was the Blue Ridge in Virginia, some 60 miles to the east. One of the explanations suggested by Price was that the boulders had been rafted to the site in the roots of trees. Dunbar and Rodgers (1957) illustrate the rafting of boulders by drift logs in their figure 88, p. 175.

ROOTING OF TREES IN ROCKY SOILS

In rocky soils and lithosols, root space is reduced with the result that tree roots are forced to develop around and between boulders and to penetrate into joints or other openings in bedrock. The rude symmetry seen in root systems of trees growing in deep soils free from mechanical obstructions is lacking in very rocky soils. Bracket-roots (large roots issuing from the root-collar of the stump) are often lacking on one or more sides. This has an important bearing on windfirmness.

Root grafting, which is coming to be recognized as a usual feature in forest stands, may be particularly common where roots are congested around boulders and in rock openings. The upturned root mass of a windthrown tree often has a net-like appearance, with emmeshed soil material and rocks. Rocks are often so clasped by roots that they remain held until the wood decays. Other rock masses, not enmeshed, are moved vertically or horizontally through the leverage of large roots or as a result of their pull when the tree is uprooted.

There is relatively little information available on the depths to which tree roots may penetrate openings in bedrock but they must be rather great. Walther (1893-1894, p. 568) mentioned tree roots, exposed in quarrying, that penetrated to depths of several meters in openings in sandstone and Vater (1927) reported pine roots at a depth of 21 feet in the same kind of rock. Rühl (1952) also observed the deep penetration of tree roots in fractures in sandstone. Near a quarry he saw a cleft 24 feet deep that contained spruce roots to the bottom. In the Rocky Mountains in Colorado roots of ponderosa

pine have been observed in fractured granite at depths approximately 35 to 40 feet below the surface. The scarcity of data on this point results primarily from lack of exposures where penetration into bedrock can be observed rather than from rare occurrence of such penetration.

UPROOTING OF TREES

The boles of trees may be viewed as cantilever beams possessing great strength and capable of exerting tremendous leverage at the point of support or fulcrum. This is particularly true of tall trees having large diameters. In general, there is a tendency for the boles of trees to conform to the requirements of beams of uniform resistance. Thus, the diameter usually increases downward and is largest at the base where stresses are greatest. Wood of higher density and greater strength is sometimes substituted for greater volume of wood. Quite generally it seems that the bole of a tree is stronger than the root system and this leads to the frequent occurrence of uprooting; breakage of boles during severe wind storms is well known but in the view of the writer is much less common than failure of the root system.

On rocky sites uprooting seems to be more common than in situations with deep soils relatively free from rocks. One reason for this is that in very rocky soils the strong bracket-roots so necessary to windfirmness may be lacking on one or more sides of the stump. A second consideration, pointed out by Mergen (1954), is that compression failures of bracket-roots commonly occur when rocks prevent the roots from moving downward during oscillation of the tree by the wind. Thus, breaks may occur in roots growing over large boulders in the soil mass.

MOVEMENT OF ROCKS

New England forests often occur on very rocky ground, with bedrock at the surface or at shallow depth. Windthrow is relatively common. Observations and measurements of rock movement by uprooted trees were made in forest stands distributed from New Hampshire to Connecticut. In 39 examples, rock masses having volumes of more than 3 cu. ft. and weights of over a quarter ton were moved vertically or horizontally distances varying from 2 to 12 feet. In two instances rocks having volumes of as much as 50 cubic feet and weights of about four and a quarter tons were moved 4 to 6 feet. Tree species concerned were mostly oaks but white pine, hemlock, yellow birch and sugar maple were also represented. Tree diameters varied from 12 to 24 inches and heights ranged from 50 to 100 feet. Rock types included granite, basalt, schist, and gneiss. With attention directed to the volume and weight of the larger rocks, no attempt was made to compute the volume and weight of the soil material moved. This commonly represented a volume of a cubic yard, or more, and a weight of at least 1,500 pounds. Plates 1 and 2 illustrate the work that may be accomplished when trees are uprooted.

Where bedrock is at or close to the surface, masses of varying size may be torn out by trees when they are uprooted. Such rock masses show fresh fracture surfaces as also does the bedrock in place from which they were broken.

SIGNIFICANCE OF ROCK MOVEMENT BY UPROOTING OF TREES

Trees perform a considerable amount of mechanical work in the zone occupied by their roots. Soil material and rocks are moved about by the roots as they grow and when trees are uprooted, usually as a result of wind. This mechanical work contributes to rock and mineral weathering and is peculiar to sites occupied by trees, particularly large trees. The process must have resulted in greater disruption and movement in the surface mantle of soil and rock in areas occupied by virgin or old-growth stands than in present-day second-growth forests. This follows from the fact that trees in the former were generally taller, larger in diameter, and possessed more massive root systems than trees in second-growth stands.

Uprooting of trees contributes to the downslope movement (creep) of the soil and rock mantle. Locally, uprooting of trees may initiate erosion of the gully type. Some of the effects of soil and rock movement by uprooting of trees are similar to those resulting from periglacial frost heaving. Increasingly, geologists and soil scientists are coming to recognize that forest trees may be important agents in weathering and in the vertical and horizontal movement of rock and soil material.

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[American Journal of Science, Vol. 258, December 1960, P. 757-768]

PROPERTIES OF WATER. PART VI. ENTROPY AND GIBBS FREE ENERGY OF WATER IN THE RANGE 10-1000°C AND 1-250,000 BARS*

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ABSTRACT. Tables of entropy and Gibbs free energy of water are presented for temperatures between 10°C and 1000°C and pressures between 1 bar and 250,000 bars. These tables have been prepared by using the thermodynamic data of Hilsenrath, et al. on steam at low pressures and by numerically differentiating and integrating the measured specific volumes of Kennedy and co-workers. The specific volumes above 2500 bars were those calculated by Knopoff et al. from a modified Thomas-Fermi-Dirac equation of state which interpolated between the shock wave data and the measured data of Kennedy, et al.

INTRODUCTION

Water is a major component in many reactions of interest to igneous, metamorphic and sedimentary petrologists. A knowledge of the thermodynamic properties of water, coupled with the thermodynamic properties of the solid phases involved in a reaction, allows one to calculate the pressures and temperatures of equilibrium. Such calculations provide an independent check on the equilibrium relations determined directly in hydrothermal experiments. Alternatively, some as yet unknown thermodynamic properties of the solid phases involved in a reaction can be obtained from experimental results of phase equilibrium studies.

AVAILABLE EXPERIMENTAL DATA

Keenan and Keyes (1936) present tables for the entropy and enthalpy of water up to about 380 bars and 871°C. These tables summarize the thermodynamical data on water available up to 1936. Osborne, Stimson and Ginnings (1939) report thermodynamical values for water along the saturation curve. Keyes (1949) confirmed the thermodynamic consistency of the data on water up to 550°C and the accuracy of the Keenan and Keyes tables. Using the specific volumes determined by Kennedy (1950), MacDonald (1955) calculated the Gibbs free energy of water for temperatures up to 800°C and pressures up to 2,500 bars.

The specific volume of water has since been extensively remeasured in the range 1-1400 bars and 0-1000°C by Kennedy and his co-workers in a series of publications (Kennedy 1957; Kennedy, Knight and Holser 1958; Holser and Kennedy 1958; Holser and Kennedy 1959). Kennedy extrapolated these results to 2,500 bars pressure (unpublished data). Knopoff, Holser and Kennedy (unpublished data) have in addition prepared tables of the specific volume of water up to 250,000 bars by use of a Thomas-Fermi-Dirac type equation of state to interpolate between the specific volumes of water at lower pressures and the specific volumes of water at these extreme pressures, as found by shock

* Publication no. 167, Institute of Geophysics, University of California, Los Angeles, California.

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TABLE 1

Molar entropy of water in range 10°C-280°C and 1-2,500 bars (cal/deg mole)

P(bar)	10°C	20°C	30°C	40°C	50°C	60°C	70°C	80°C	90°C	100°C	120°C	140°C	160°C	180°C	200°C	220°C	240°C	260°C	280°C
1	15.817	16.443	17.050	17.634	18.200	18.749	19.282	19.802	20.307	20.800	47.253	47.679	48.086	48.472	48.839	49.191	49.531	49.857	50.175
10	15.817	16.442	17.049	17.632	18.198	18.747	19.280	19.799	20.304	20.797	21.731	22.693	23.590	43.630 24.507	43.975	44.350	44.685	45.110	45.460
50	15.816	16.439	17.043	17.625	18.190	18.738	19.270	19.788	20.292	20.784	21.716	22.675	23.570	24.484	25.267	25.990	26.823	27.566	41.240
100	15.814	16.434	17.036	17.617	18.180	18.726	19.257	19.774	20.277	20.767	21.697	22.653	23.545	24.455	25.234	25.951	26.779	27.511	28.285
150	15.811	16.430	17.029	17.608	18.170	18.715	19.245	19.760	20.262	20.751	21.678	22.631	23.520	24.427	25.202	25.914	26.735	27.457	28.196
200	15.809	16.425	17.022	17.600	18.160	18.704	19.232	19.747	20.247	20.735	21.659	22.610	23.496	24.400	25.171	25.878	26.692	27.406	28.134
250	15.806	16.420	17.016	17.591	18.150	18.693	19.220	19.733	20.233	20.719	21.640	22.588	23.472	24.373	25.140	25.843	26.652	27.358	28.077
300	15.803	16.415	17.009	17.583	18.140	18.681	19.207	19.719	20.218	20.704	21.622	22.568	23.448	24.347	25.111	25.810	26.613	27.312	28.020
350	15.799	16.410	17.002	17.574	18.130	18.670	19.195	19.706	20.203	20.688	21.604	22.547	23.426	24.321	25.082	25.777	26.575	27.268	27.969
400	15.796	16.405	16.995	17.566	18.120	18.659	19.183	19.693	20.189	20.673	21.586	22.527	23.403	24.296	25.054	25.745	26.539	27.226	27.919
450	15.792	16.400	16.988	17.557	18.110	18.648	19.171	19.680	20.174	20.657	21.569	22.508	23.381	24.271	25.026	25.714	26.503	27.185	27.873
500	15.788	16.394	16.981	17.549	18.100	18.637	19.159	19.666	20.161	20.642	21.552	22.489	23.360	24.247	25.000	25.684	26.469	27.146	27.828
600	15.780	16.383	16.967	17.532	18.081	18.616	19.135	19.641	20.134	20.613	21.518	22.451	23.317	24.200	24.947	25.626	26.403	27.071	27.744
700	15.771	16.372	16.952	17.514	18.061	18.594	19.112	19.615	20.106	20.584	21.485	22.414	23.277	24.155	24.900	25.570	26.341	27.001	27.662
800	15.761	16.360	16.937	17.497	18.041	18.572	19.089	19.590	20.079	20.555	21.454	22.378	23.237	24.112	24.849	25.517	26.282	26.935	27.587
900	15.751	16.348	16.922	17.480	18.022	18.551	19.066	19.565	20.053	20.527	21.422	22.344	23.199	24.070	24.803	25.466	26.226	26.872	27.516
1,000	15.740	16.335	16.907	17.462	18.003	18.530	19.043	19.541	20.027	20.500	21.392	22.310	23.162	24.029	24.758	25.416	26.171	26.812	27.451
1,100	15.729	16.322	16.891	17.444	17.983	18.510	19.021	19.517	20.001	20.473	21.362	22.277	23.126	23.989	24.714	25.369	26.119	26.754	27.386
1,200	15.717	16.309	16.876	17.427	17.964	18.489	18.999	19.493	19.976	20.447	21.333	22.244	23.090	23.951	24.672	25.323	26.069	26.699	27.316
1,300	15.705	16.295	16.860	17.409	17.945	18.469	18.977	19.469	19.951	20.421	21.304	22.213	23.056	23.913	24.611	25.278	26.020	26.647	27.257
1,400	15.692	16.281	16.844	17.392	17.926	18.449	18.956	19.446	19.926	20.395	21.276	22.182	23.022	23.877	24.592	25.235	25.974	26.596	27.202
1,600	15.666	16.252	16.811	17.357	17.890	18.409	18.913	19.399	19.878	20.346	21.221	22.123	22.958	23.806	24.515	25.153	25.885	26.498	27.099
1,800	15.638	16.219	16.776	17.322	17.855	18.371	18.870	19.354	19.832	20.300	21.168	22.067	22.897	23.740	24.442	25.074	25.801	26.407	27.002
2,000	15.608	16.185	16.742	17.288	17.821	18.332	18.827	19.311	19.789	20.255	21.118	22.014	22.840	23.676	24.373	25.000	25.721	26.319	26.902
2,500	15.533	16.099	16.656	17.202	17.735	18.235	18.719	19.204	19.682	20.146	21.002	21.896	22.710	23.531	24.217	24.830	25.538	26.118	26.673

wave methods. For temperatures below 100°C Bridgman's specific volume data (1912, 1931) are also available.

METHODS OF CALCULATION

Since an increasing amount of experimentation is being carried out at very high water pressures, and since many geologically important reactions may take place at these pressures, it is desirable to extend the presently available tables of thermodynamic data of water to high pressures by using the measured and calculated specific volumes of Kennedy and his group.

The Keenan and Keyes tables, as well as the Hilsenrath tables, provide data for the entropy and Gibbs free energy of steam at 1 bar over a range of temperatures. Enthalpies of evaporation are given in the Keenan and Keyes tables, and the heat capacities of liquid water in the range 1-100°C are given by Osborne, Stimson and Ginnings (1939).

This information plus the specific volume measurements can be combined to give the desired thermochemical data. The equations used are

$$S = S_o - \int_{P_o}^{P} \left(\partial V / \partial T \right)_P dP$$

and

$$G = G_o + \int_{P_o}^{P} V dP$$

where S and G are the desired entropy and Gibbs free energy, S_o and G_o are the entropy and Gibbs free energy at the given starting pressure P_o and V is the volume. Evaluation of derivatives from the tabulated values is somewhat unsatisfactory, in that it depends on the differentiation of experimental data, which are affected in an unpredictable way by rounding errors and random experimental errors. Differentiation tends to exaggerate such errors. However, these difficulties are not circumvented by fitting en equation of state to the experimental data, since an equation with a number of parameters sufficient to do justice to the data will also tend to reproduce the irregularities.

The present calculations were carried out by fitting a set of nine experimental points to the equation

$$V = (a_1 + a_2 T + a_3 T^2) + (b_1 + b_2 T + b_3 T^2) P + (c_1 + c_2 T + c_3 T^2)/P.$$

The derivative $\left(\partial V / \partial T \right)_P$ and the integrals for ΔS and ΔG were evaluated from this equation. The fitting of the above equation was repeated for each increment in pressure, until an individual isotherm had been completed. These calculations were carried out on a Bendix G-15 digital computer.

In the calculation of G the experimental values are integrated. Rounding and nonsystematic experimental errors tend to be reduced, although systematic errors remain.

Kelley's (1950) recommended value for the entropy of water of 16.75 ± 0.03 cal/deg mole at 25°C is used. *The zero reference point of the free energy function is taken as the internal energy of the ideal gas at absolute zero*, as in the Hilsenrath tables (1955).

TABLE 2

Molar entropy of water in the range 300°-1000° and 1-2500 bars (cal/deg mole)

P(Bar)	300°C	320°C	340°C	360°C	380°C	400°C	420°C	440°C	460°C	480°C
1	50.481	50.781	51.069	51.351	51.627	51.893	52.156	52.410	52.663	52.907
10	45.790	46.120	46.410	46.710	46.990	47.278	47.541	47.776	48.112	48.448
20	44.334	44.558	44.863	45.235	45.471	45.799	46.121	46.360	46.700	47.040
30	43.316	43.581	43.910	44.271	44.552	44.858	45.171	45.430	45.789	46.148
40	42.526	42.833	43.174	43.594	43.843	44.183	44.512	44.784	45.156	45.528
50	41.850	42.197	42.561	43.028	43.294	43.674	43.998	44.258	44.618	44.979
60	41.232	41.661	42.053	42.557	42.819	43.198	43.565	43.831	44.197	44.563
70	40.623	41.139	41.585	42.105	42.384	42.794	43.181	43.445	43.810	44.175
80	40.009	40.629	41.121	41.678	41.978	42.433	42.854	43.128	43.502	43.876
90	29.090	40.114	40.677	41.295	41.611	42.111	42.531	42.803	43.176	43.549
100	29.078	39.606	40.236	40.913	41.263	41.789	42.251	42.527	42.904	43.281
150	28.985	29.776	30.806	38.959	39.580	40.406	40.895	41.257	41.719	42.181
200	28.903	29.650	30.579	32.078	—	39.116	39.674	40.140	40.706	41.273
250	28.827	29.546	30.402	31.506	—	36.630	38.433	39.040	39.747	40.455
300	28.759	29.452	30.262	31.202	—	34.365	36.536	37.501	38.567	39.633
350	28.696	29.368	30.145	31.010	—	33.288	35.099	36.270	37.541	38.813
400	28.638	29.291	30.041	30.857	—	32.757	34.152	35.377	36.703	38.029
450	28.582	29.220	29.951	30.730	—	32.469	33.593	34.725	35.957	37.190
500	28.529	29.155	29.868	30.618	—	32.252	33.247	34.255	35.364	36.473
600	28.428	29.038	29.719	30.436	—	31.927	32.774	33.620	34.566	35.512
700	28.334	28.927	29.590	30.288	—	31.685	32.451	33.185	34.020	34.855
800	28.251	28.829	29.478	30.158	—	31.485	32.205	32.876	33.647	34.419
900	28.174	28.740	29.374	30.035	—	31.320	32.000	32.626	33.352	34.078
1000	28.096	28.651	29.279	29.930	—	31.164	31.830	32.423	33.116	33.810
1100	28.029	28.576	29.188	29.827	—	31.036	31.686	32.251	32.916	33.582
1200	27.963	28.511	29.104	29.730	—	30.923	31.565	32.109	32.754	33.399
1300	27.898	28.443	29.026	29.643	—	30.816	31.443	31.973	32.604	33.235
1400	27.834	28.372	28.952	29.556	—	30.712	31.332	31.851	32.470	33.089
1600	27.721	28.246	28.814	29.401	—	30.517	31.143	31.648	32.254	32.860
1800	27.611	28.125	28.684	29.257	—	30.349	30.959	31.455	32.052	32.649
2000	27.510	28.021	28.579	29.125	—	30.190	30.796	31.287	31.878	32.470
2500	27.278	27.784	28.343	28.840	—	29.837	30.430	30.920	31.510	32.101

TABLE 2 (Continued)

500°C	550°C	600°C	650°C	700°C	750°C	800°C	850°C	900°C	950°C	1000°C
53.147	53.728	54.278	54.811	55.321	55.814	56.291	56.754	57.201	57.634	58.058
48.635	49.157	49.695	50.209	50.754	51.218	51.682	52.146	52.644	53.090	53.626
47.221	47.753	48.302	48.820	49.359	49.818	50.277	50.737	51.278	51.706	52.236
46.370	46.923	47.470	47.986	48.532	48.993	49.455	49.917	50.460	50.875	51.410
45.760	46.316	46.872	47.391	47.937	48.390	48.858	49.319	49.875	50.298	50.822
45.275	45.837	46.399	46.922	47.475	47.937	48.395	48.862	49.422	49.843	50.375
44.876	45.442	46.008	46.537	47.092	47.556	48.018	48.482	49.044	49.473	50.002
44.534	45.103	45.672	46.207	46.768	47.232	47.696	48.160	48.722	49.160	49.681
44.227	44.802	45.377	45.914	46.489	46.951	47.414	47.877	48.445	48.878	49.398
43.954	44.532	45.110	45.654	46.235	46.699	47.163	47.627	48.201	48.635	49.156
43.703	44.286	44.869	45.414	46.002	46.491	46.980	47.470	47.979	48.419	48.940
42.661	43.280	43.899	44.467	45.095	45.577	46.059	46.541	47.114	47.555	48.097
41.829	42.493	43.157	43.759	44.412	44.912	45.412	45.913	46.494	46.928	47.472
41.100	41.816	42.532	43.174	43.863	44.380	44.901	45.420	46.010	46.443	46.978
40.424	41.206	41.988	42.669	43.389	43.926	44.459	44.994	45.603	46.037	46.566
39.787	40.638	41.489	42.218	42.973	43.523	44.073	44.624	45.245	45.679	46.206
39.181	40.105	41.029	41.802	42.592	43.161	43.730	44.300	44.931	45.369	45.891
38.598	39.597	40.596	41.417	42.247	42.834	43.421	44.009	44.653	45.084	45.608
38.038	39.112	40.186	41.058	41.924	42.530	43.136	43.742	44.392	44.828	45.349
37.033	38.234	39.435	40.396	41.329	41.972	42.616	43.260	43.925	44.374	44.889
36.263	37.519	38.775	39.803	40.796	41.474	42.153	42.832	43.513	43.967	44.488
35.695	36.951	38.207	39.276	40.323	41.031	41.740	42.449	43.149	43.614	44.131
35.268	36.497	37.726	38.814	39.898	40.631	41.364	42.098	42.815	43.288	43.805
34.938	36.129	37.320	38.413	39.520	40.272	41.024	41.776	42.510	42.985	43.515
34.673	35.827	36.981	38.064	39.183	39.950	40.715	41.482	42.232	42.717	43.249
34.462	35.568	36.674	37.758	38.887	39.662	40.438	41.214	41.976	42.456	42.990
34.228	35.346	36.464	37.493	38.592	39.379	40.167	40.954	41.729	42.216	42.755
34.032	35.152	36.272	37.244	38.315	39.115	39.915	40.716	41.499	41.989	42.530
33.763	34.831	35.899	36.837	37.890	38.691	39.492	40.294	41.084	41.586	42.137
33.544	34.570	35.596	36.505	37.537	38.332	39.126	39.921	40.723	41.233	41.768
33.360	34.354	35.348	36.234	37.244	38.028	38.812	39.597	40.407	40.918	41.450
32.985	33.929	34.873	35.736	36.711	37.450	38.190	38.930	39.752	40.282	40.834

TABLE 3

Molar entropy of water in range 10°C-180°C and 3,000-25,000 bars (cal/deg mole)

P(Bar)	10°C	20°C	30°C	40°C	50°C	60°C	70°C	80°C	90°C	100°C	160°C	180°C
3,000	15.458	16.018	16.570	17.116	17.643	18.138	18.617	19.096	19.574	20.036	—	—
3,500	15.377	15.943	16.489	17.030	17.546	18.041	18.526	18.994	19.467	19.927	—	—
4,000	15.291	15.869	16.414	16.943	17.456	17.950	18.434	18.892	19.359	19.819	—	—
5,000	15.130	15.718	16.263	16.771	17.283	17.778	18.241	18.687	19.155	19.608	22.185	22.858
6,000	14.990	15.556	16.102	16.599	17.111	17.606	18.059	18.504	18.972	19.414	—	—
7,000	—	15.384	15.930	16.427	16.938	17.433	17.896	18.343	18.800	19.235	—	—
8,000	—	—	15.757	16.255	16.766	17.261	17.746	18.192	18.627	19.055	—	—
9,000	—	—	—	16.083	16.594	17.089	17.595	18.042	18.455	18.876	—	—
10,000	—	—	—	15.911	16.422	16.917	17.434	17.880	18.283	18.700	21.351	—
15,000	—	—	—	—	—	—	16.627	17.073	17.476	17.947	20.706	—
20,000	—	—	—	—	—	—	—	—	16.831	17.409	20.221	—
25,000	—	—	—	—	—	—	—	—	—	—	19.791	—

TABLE 4

Molar entropy of water in range 200°C-1,000°C and 5,000-250,000 bars (cal/deg mole)

P(Bar)	200°C	300°C	400°C	500°C	750°C	1000°C
5,000	23.639	26.458	28.813	31.807	35.823	38.712
10,000	22.886	25.535	27.757	30.624	34.485	37.141
15,000	22.240	24.911	27.091	29.918	33.754	36.364
20,000	21.648	24.407	26.588	29.398	33.223	35.820
25,000	21.164	23.961	26.146	28.949	32.772	35.365
30,000	20.734	23.542	25.728	28.527	32.348	34.942
40,000	—	22.746	24.926	27.724	31.544	34.139
50,000	—	21.992	24.172	26.971	30.796	33.392
100,000	—	18.943	21.103	23.920	27.776	30.337
150,000	—	16.648	18.915	21.818	25.630	28.176
200,000	—	15.246	17.457	20.297	24.097	26.643
250,000	—	14.220	16.401	19.216	23.008	25.554

No values are listed for the entropy of water at pressures above 200 bars in the case of the 380°C isotherm. This is due to the large uncertainties in volumes and thermal data in the immediate region of the critical point.

Tables 1, 2, 3 and 4 list values of the molar entropy of water for temperatures ranging from 10°C to 1000°C and pressures ranging from 1 bar to 250,000 bars. Tables 5, 6, 7 and 8 list values of the negative molar Gibbs free energy of water for the same range of pressures and temperatures.

ACCURACY OF VALUES

It is extremely difficult to make a reliable estimate of the accuracy of the values of entropy and Gibbs free energy listed in tables 1-8. Estimates of the accuracy of the 1 bar values at different temperatures are given elsewhere (Keyes 1949; Wagman, et al. 1945; Kelley 1950; Hilsenrath, et al. 1955). It is believed that the additional error due to random errors in the experimental data and to the method of calculation is of the order of 2% of the difference between the free energy at P and the free energy at 1 bar, for any given temperature. Above 2500 bars this estimate should be doubled. The same additional error in the case of the entropies may be taken as at least 6 percent. Above 200 bars the 400°C isotherm may be in error to an unknown, but large, amount, especially for the entropies, due to the proximity of the critical point.

ACKNOWLEDGMENTS

The authors wish to thank Drs. George C. Kennedy and Gordon J. F. MacDonald for their interest in this project. Thanks are owing to the office of Naval research for partial financial support of this effort under contract NONR 233C27.

TABLE 5

Negative molar Gibbs free energy of water in the range 10°C-280°C and 1-2500 bars (cal/mole)

P(Bar)	10°C	20°C	30°C	40°C	50°C	60°C	70°C	80°C	90°C	100°C	120°C	140°C	160°C	180°C	200°C	220°C	240°C	260°C	280°C
1	12889	13050	13217	13391	13570	13755	13945	14140	14341	14546	15487	16436	17394	18359	19333	20312	21299	22294	23294
10	12885	13046	13213	13387	13566	13751	13941	14136	14337	14542	14963	15398	15861	16320	17200	18062	18972	19814	20798
50	12868	13029	13196	13370	13549	13734	13923	14118	14319	14524	14945	15379	15842	16301	16794	17309	17831	18293	19160
100	12846	13007	13174	13348	13527	13712	13902	14096	14297	14502	14922	15356	15818	16277	16769	17285	17805	18345	18905
150	12825	12986	13153	13327	13505	13690	13880	14074	14275	14479	14900	15333	15795	16253	16745	17258	17779	18318	18877
200	12804	12965	13131	13305	13484	13668	13858	14052	14253	14457	14877	15310	15771	16229	16720	17233	17753	18291	18849
250	12782	12943	13110	13284	13462	13647	13836	14030	14231	14435	14854	15287	15748	16205	16696	17208	17727	18264	18821
300	12761	12922	13089	13262	13441	13625	13814	14008	14209	14413	14832	15264	15725	16181	16671	17183	17701	18238	18793
350	12740	12901	13067	13241	13419	13603	13793	13987	14187	14391	14809	15241	15701	16157	16647	17158	17675	18211	18766
400	12719	12880	13046	13219	13398	13582	13771	13965	14165	14368	14787	15218	15678	16134	16623	17133	17650	18185	18739
450	12698	12858	13025	13198	13376	13560	13749	13943	14143	14346	14765	15196	15655	16110	16599	17108	17625	18159	18712
500	12677	12837	13004	13177	13355	13539	13728	13921	14121	14325	14742	15173	15632	16087	16574	17084	17599	18133	18685
600	12635	12795	12961	13135	13312	13496	13685	13878	14077	14281	14698	15128	15586	16040	16527	17035	17549	18081	18632
700	12593	12753	12919	13092	13270	13454	13642	13835	14034	14237	14654	15083	15540	15993	16479	16986	17499	18030	18579
800	12551	12711	12877	13050	13228	13411	13599	13792	13991	14194	14610	15038	15495	15947	16432	16938	17450	17980	18527
900	12510	12670	12836	13008	13186	13369	13557	13749	13948	14150	14566	14994	15450	15901	16385	16890	17401	17929	18476
1,000	12468	12628	12794	12967	13144	13327	13514	13707	13905	14107	14522	14949	15404	15855	16338	16842	17352	17880	18425
1,100	12427	12587	12753	12925	13102	13285	13472	13664	13862	14064	14479	14905	15360	15809	16292	16795	17304	17830	18374
1,200	12386	12546	12711	12883	13060	13243	13430	13622	13820	14021	14435	14861	15315	15764	16246	16748	17256	17781	18324
1,300	12345	12505	12670	12842	13019	13201	13388	13580	13777	13979	14392	14817	15271	15719	16200	16701	17208	17732	18274
1,400	12304	12464	12629	12801	12977	13159	13346	13538	13735	13936	14349	14774	15226	15674	16154	16655	17161	17684	18225
1,600	12223	12383	12548	12719	12895	13077	13263	13454	13651	13852	14263	14687	15138	15584	16063	16562	17067	17588	18127
1,800	12143	12302	12466	12637	12813	12994	13181	13371	13568	13768	14178	14601	15051	15496	15973	16471	16974	17493	18030
2,000	12063	12221	12386	12556	12732	12913	13098	13289	13484	13684	14094	14515	14964	15408	15884	16380	16881	17399	17934
2,500	11864	12022	12186	12355	12530	12710	12895	13084	13279	13477	13885	14304	14750	15191	15664	16157	16655	17169	17700

TABLE 6

Negative molar Gibbs free energy of water in the range 300°C-1000°C and 1-2500 bars (cal/mole)

P(Bar)	300°C	320°C	340°C	360°C	380°C	400°C	420°C	440°C	460°C	480°C
1	24301	25313	26331	27356	28386	29421	30462	31508	32558	33614
10	21721	22621	23559	24479	25500	26360	27312	28264	29216	30169
20	20953	21806	22689	23615	24598	25454	26386	27318	28250	29181
30	20505	21345	22205	23115	24078	24927	25830	26733	27636	28541
40	20198	21031	21878	22780	23719	24551	25443	26335	27227	28120
50	19960	20780	21619	22514	23456	24280	25156	26032	26908	27783
60	19784	20592	21419	22312	23241	24054	24920	25786	26702	27518
70	19634	20437	21255	22140	23057	23870	24725	25594	26491	27297
80	19511	20312	21123	21987	22900	23709	24543	25411	26171	27106
90	19445	20207	21008	21867	22766	23571	24402	25265	26012	26938
100	19440	20117	20910	21758	22651	23439	24276	25134	25875	26795
150	19410	20002	20598	21384	22219	22957	23779	24669	25419	26255
200	19380	19970	20561	21219	21967	22695	23453	24348	25087	25931
250	19350	19939	20526	21178	21789	22484	23261	24108	24851	25669
300	19322	19909	20491	21140	21742	22400	23118	23953	24638	25466
350	19294	19878	20459	21105	21717	22374	23054	23837	24532	25338
400	19266	19848	20426	21069	21680	22312	23020	23745	24458	25239
450	19238	19818	20393	21035	21644	22272	22977	23655	24387	25151
500	19211	19788	20363	21003	21609	22235	22934	23603	24326	25045
600	19156	19731	20303	20938	21541	22162	22851	23505	24219	24906
700	19102	19673	20242	20876	21476	22090	22772	23425	24127	24800
800	19049	19616	20185	20814	21414	22022	22700	23346	24047	24699
900	18997	19560	20129	20753	21351	21958	22631	23274	24014	24612
1000	18943	19506	20070	20693	21290	21892	22563	23203	23892	24528
1100	18892	19453	20014	20635	21227	21832	22500	23137	23830	24448
1200	18841	19402	19957	20578	21170	21773	22437	23069	23751	24373
1300	18790	19349	19901	20521	21113	21714	22375	23005	23686	24299
1400	18739	19295	19848	20466	21055	21655	22314	22945	23624	24227
1600	18641	19192	19740	20356	20948	21537	22192	22821	23501	24095
1800	18544	19089	19628	20245	20835	21424	22074	22702	23380	23967
2000	18446	18989	19527	20136	20727	21311	21960	22587	23264	23830
2500	18206	18745	19280	19870	20464	21036	21684	22308	22986	23502

TABLE 6 (Continued)

500°C	550°C	600°C	650°C	700°C	750°C	800°C	850°C	900°C	950°C	1000°C
34674	37347	40041	42769	45522	48303	51105	53932	56783	59649	62544
31113	33606	36027	38539	41054	43622	46209	48798	51409	54140	56829
30074	32487	34832	37273	39714	42217	44739	47252	49818	52455	55072
29460	31839	34139	36538	38944	41395	43877	46348	48868	51484	54057
29029	31380	33646	36019	38395	40818	43268	45707	48217	50792	53331
28695	31026	33268	35615	37970	40374	42799	45210	47704	50244	52780
28425	30738	32960	35289	37623	40010	42412	44805	47275	49805	52315
28195	30497	32701	35014	37330	39702	42087	44462	46916	49441	51922
28002	30289	32478	34777	37080	39436	41808	44165	46609	49117	51580
27833	30108	32283	34567	36858	39199	41560	43906	46348	48833	51282
27681	29945	32109	34380	36661	38992	41341	43674	46107	48583	51022
27111	29335	31451	33676	35920	38196	40498	42786	45173	47609	50001
26744	28917	30995	33183	35392	37642	39906	42161	44517	46924	49289
26470	28605	30652	32810	34988	37213	39452	41684	44015	46399	48738
26260	28361	30381	32513	34665	36867	39080	41293	43608	45988	48281
26092	28164	30157	32266	34396	36582	38771	40963	43261	45620	47901
25959	28001	29969	32055	34165	36334	38504	40682	42963	45307	47566
25850	27863	29809	31873	33967	36117	38270	40436	42702	45034	47277
25761	27746	29669	31716	33791	35927	38063	40216	42471	44789	47018
25607	27554	29438	31449	33498	35604	37710	39840	42072	44375	46564
25488	27403	29251	31231	33251	35335	37417	39518	41734	44015	46182
25391	27276	29094	31047	33040	35103	37163	39245	41440	43706	45850
25300	27164	28957	30889	32862	34901	36940	39007	41190	43433	45558
25214	27063	28835	30747	32702	34723	36744	38793	40966	43188	45298
25133	26970	28724	30620	32560	34564	36566	38603	40760	42973	45068
25057	26882	28623	30501	32430	34417	36407	38428	40569	42767	44848
24983	26799	28510	30403	32319	34280	36258	38266	40393	42577	44649
24911	26720	28419	30298	32201	34153	36119	38116	40233	42404	44460
24772	26569	28252	30111	31993	33922	35867	37841	39936	42083	44122
24640	26427	28095	29935	31803	33715	35643	37595	39675	41806	43824
24513	26291	27946	29774	31628	33525	35434	37373	39436	41552	43557
24207	25970	27601	29404	31233	33103	34977	36888	38917	41002	42981

TABLE 7

Negative molar Gibbs free energy of water in the range 10°C-200°C and 3,000-30,000 bars (cal/mole)

P(Bar)	10°C	20°C	30°C	40°C	50°C	60°C	70°C	80°C	90°C	100°C	160°C	180°C	200°C
3,000	11669	11826	11988	12157	12331	12510	12694	12882	13075	13273	—	—	—
3,500	11475	11632	11793	11961	12134	12312	12495	12682	12875	13071	—	—	—
4,000	11284	11440	11601	11768	11940	12117	12299	12485	12677	12872	—	—	—
5,000	10908	11062	11222	11387	11557	11733	11913	12097	12287	12480	13722	14151	14613
6,000	10539	10691	10849	11013	11182	11356	11534	11716	11904	12095	—	—	—
7,000	10176	10327	10484	10646	10813	10985	11161	11342	11528	11718	—	—	—
8,000	—	9969	10124	10284	10449	10619	10794	10974	11158	11346	—	—	—
9,000	—	—	9769	9928	10091	10259	10433	10611	10793	10980	—	—	—
10,000	—	—	9418	9575	9737	9904	10075	10252	10433	10617	11800	12212	12659
15,000	—	—	—	—	—	8181	8344	8513	8686	8863	10004	10404	10838
20,000	—	—	—	—	—	—	—	6852	7017	7188	8300	8690	9112
25,000	—	—	—	—	—	—	—	—	—	—	6665	7046	7458
30,000	—	—	—	—	—	—	—	—	—	—	5083	5456	5859

TABLE 8

Negative molar Gibbs free energy of water in range 300°C-1,000°C and 5,000-250,000 bars (cal/mole)

P(Bar)	300°C	400°C	500°C	750°C	1000°C
5,000	17091	19826	22887	31438	40848
10,000	15054	17694	20640	28885	37932
15,000	13173	15751	18627	26695	35554
20,000	11397	13926	16750	24688	33412
25,000	9699	12184	14963	22790	31401
30,000	8059	10501	13239	20959	29465
40,000	4912	7275	9933	17451	25756
50,000	1914	4202	6785	14116	22234
100,000	−11430	−9452	−7179	−593	6760
150,000	−23077	−21325	−19264	−13210	−6396
200,000	−33723	−32106	−30198	−24529	−18093
250,000	−43739	−42220	−40422	−35022	−28860

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[American Journal of Science, Vol. 258, December 1960, P. 769-772]

OCCURRENCE OF CuCl EMISSION IN VOLCANIC FLAMES*

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ABSTRACT. Four molecular bands recorded in spectrograms taken of volcanic flames during the 1960 eruption of Kilauea prove to be the strongest radiation bands of the molecule CuCl. A comparison of the Cu/Na ratio in the flame and in a previously described Kilauea sublimate suggests that an appreciable part of the copper that is volatized out of Hawaiian magmas is transported as CuCl.

INTRODUCTION

Many investigators have made qualitative, visual spectroscopic observations on volcanic flames, but Verhoogen (1939) first placed the study on a quantitative basis by obtaining spectrograms of the flames during the 1938 eruption of the basaltic volcano Nyamlagira. Among the emitting atoms and molecules identified in his spectrograms were Na, K, and N_2. Such spectrograms do not disclose the nature of the major burning gases, apparently because the molecules and radicals involved in the combustion are poor emitters of radiation, at least in the spectral region examined. Rather, colors of volcanic flames seem to be due more to minor components that emit their characteristic radiation strongly under the prevailing thermal and physical-chemical conditions. These minor components offer a means of determining some aspects of the chemistry and energetics of volcanic flames and thereby comprise a very interesting subject for continued study.

PRESENT WORK

During the 1960 flank eruption of Kilauea Volcano, flames were seen to be issuing intermittently for only a few days (February 7 to 10) from a subsidiary vent on the west side of the main group of cones near the village of Kapoho, Hawaii. The flames varied in color from yellowish white to yellow green, were frequently diluted by white steam, and were contaminated by thin wisps of molten ash and pumice expelled from the vent.

Spectrograms of the flames were obtained at night by means of a small Gaertner quartz spectrograph loaded with Eastman 103aF plate. The image of the flame was focussed on the slit of the spectrograph by means of a quartz condensing lens with a focal length of 14 cm, and exposures varying between 15 seconds and 4.5 hours were tried. A comparison spectrum for wavelength determination was exposed above each flame spectrum by means of a helium Geissler tube activated by a spark coil and dry cells. Owing to wide variations in the intensity of the flames, only four measurable spectrograms were obtained. In retrospect, a glass spectrograph offering greater dispersion in the visible region would have been preferable because no radiation shorter than 370 millimicrons was recorded even in the darkest spectrograms.

The spectrograms were examined with a Gaertner comparator, and calibration curves relating distance on the plate to wavelength were constructed

* Publication authorized by the Director, U. S. Geological Survey.

from measurements of lines in the spectra of helium. As indicated in table 1, only the yellow D lines of sodium, four strong bands assignable to CuCl, and four faint bands of uncertain origin were found. The compilation by Pearse and Gaydon (1950) was used to identify the bands.

TABLE 1

Wavelength and identity of bands and lines in spectra of Kilauea flames

λ in flame	λ (Pearse and Gaydon, 1950)	Emitter	Remarks
mμ	mμ		
589.5	589.3 (avg.)	Na	Unresolved doublet 589.59 and 589.00
449.5	449.4	CuCl	System E
443.0	443.4	CuCl	System D broadened by 441.2 of system E
435.5	435.4	CuCl	System D broadened by 433.3 of system E
428.5	428.1	CuCl	System D broadened by 425.9 of system E
402.5	(?)	(?)	Faint
393.5	393.9 (?)	S_2 (?)	Faint
383.0	383.7 (?)	S_2 (?)	Faint
370.0	(?)	(?)	Faint

In order to confirm the identity of the four strong bands (449.5 to 428.5 mμ), a flame rich in CuCl emission was artificially produced in the laboratory by inserting a piece of asbestos cloth wet with cupric chloride solution into the flame of a bunsen burner. Cupric chloride is decomposed to cuprous chloride in the flame. The resulting flame was recorded with the same spectrograph and same kind of plate as had been used in the field. The CuCl bands of D and E systems recorded in this manner well matched those in the spectrograms of volcanic flames as to wavelength, relative intensity, and general appearance, leaving no doubt as to the correctness of the identification. The weaker bands of the A, B, and C systems of CuCl were not detected in the spectrograms of volcanic flames, however, being either unrecorded in the lighter exposures or blotted out by continuous radiation in the darker exposures.

Four very faint bands, 402.5, 393.5, 383.0, and 370.0mμ, were recorded on the best spectrogram. Two of these, 393.5 and 383.0 mμ, agree closely with published wavelengths for strong bands of the diatomic molecule S_2, but the identification must remain provisional. Shepherd's analyses (1938) of Kilauea gases show a content of S_2 varying between 0.00 and 8.61 volume percent with most samples containing tenths of percent. No satisfactory identification could be made for the other two bands, 402.5 and 370.0 mμ. The four faint bands are listed in table 1 mainly so that future workers might clarify their identity in better spectrograms.

Verhoogen (1939) reported three bands, 436.0, 443.0, and 444.0 mμ, of uncertain origin which appeared in only a few of his early spectrograms. It is interesting that the wavelengths of the first two of these are very close to those of strong CuCl bands and thereby raise the possibility that CuCl was also present in the volcanic flames of Nyamlagira. The several bands of the first positive system of N_2 reported by Verhoogen were not detected in the present study.

DISCUSSION OF RESULTS

The occurrence of CuCl in the flames of Hawaiian volcanoes might have been expected on several grounds. Copper is a relatively abundant minor element in basaltic magmas (Goldschmidt, 1954), and small patches of blue copper minerals are common in Hawaiian lavas. Chlorine or chloride has been determined in moderate amounts in Kilauea gases (Shepherd, 1938). CuCl is an efficient emitter of its characteristic radiation in flames and would color flames even at low concentrations. Its use in qualitative flame tests for copper is well known (Hodgman, 1957). Thus the availability of the elements and the emission characteristics of the molecule both favor the presence and detection of CuCl in flames of basaltic volcanoes.

The absolute concentration of CuCl in the Kilauea flames cannot be determined from the data on hand. However, the concentration relative to sodium may be roughly estimated from the relative intensities of the 435.4 mμ band of CuCl and of the D lines of sodium in the spectrograms. A rough scale of intensities was first constructed by visually estimating the darkness of the lines in the comparison spectrum of helium. The visual estimates were then refined by comparing them with the two sets of measured intensities of helium lines listed by Crosswhite and Dieke (1957), and the set designated I_2 by them was finally adopted as the reference intensity scale. The intensity of the 435.4 mμ band of CuCl in the flame is approximately 1/70 of that of the sodium D lines. The excitation potentials of CuCl and of Na are 2.84 and 2.10 electron-volts respectively, so the radiation efficiency of the two substances in the flame might be expected to be about the same. If the radiation efficiencies were the same, the molecular ratio CuCl/Na would also be 1/70. The weight ratio of the elements, Cu/Na, would then be around 1/25.

Sublimates, particularly of alkali sulfates, are commonly observed in cracks, tubes, and other openings in freshly erupted Hawaiian lavas and pumice deposits (Jaggar, 1920). Washington and Merwin (1921) described such a sublimate that was formed during the 1920 eruption of Kilauea, and their analysis showed 52.12 percent Na_2SO_4 and 0.94 percent $CuSO_4$, along with sulfates of potassium and calcium. The weight ratio Cu/Na in the sublimate calculates to 1/45.9, which is the same order of magnitude of the ratio deduced above for these elements in the flame of the 1960 eruption of Kilauea. This agreement in the order of magnitude of Cu/Na ratios between a flame and a sublimate suggests that a substantial part of the copper that is volatilized moves as CuCl.

Washington and Merwin (1921) found only 0.03 percent of Cl in the sublimate, far less than the amount equivalent to the copper. But the sulfuric acid that is formed by the oxidation of SO_2 in the cooler, air-contaminated en-

vironment of deposition may have displaced the chloride from the sublimate, leaving only the sulfate of copper. Action of rainwater on primary sublimates of copper produces secondary minerals such as brochantite that are found in the older lavas of Hawaii (Stone, 1925).

Some evidence against the volatilization of copper as the chloride has been adduced in Krauskopf's thermodynamical calculations (1959) concerning the heavy metal content of magmatic vapor, and in observations by Zies (1929) on fumarolic minerals of the Katmai region. Both of these studies agree that in vapors having temperatures around 600°C and containing appreciable amounts of HCl and H_2S there would be negligible transport of copper in the form of CuCl.

Magmatic vapors of basaltic volcanoes have temperatures (1000° to 1200°C) much higher than 600°C, may contain chlorine as Cl_2 or NaCl, and have more SO_2 than H_2S (Shepherd, 1938; Ellis, 1957). Their physicochemical state is thus quite different from those considered by Krauskopf and observed by Zies. Consequently, the conclusion reached by these investigators regarding the nonvolatility of CuCl may not hold for vapors emanating from basaltic magmas. The results of the present study strongly suggest that much of the copper volatized out of Hawaiian basaltic magmas is transported as CuCl.

ACKNOWLEDGMENT

The help and encouragement received from my colleague, J. P. Eaton, during the course of this study are gratefully acknowledged.

Note added in proof: The discovery of CuCl in the flames of Nyiragongo Volcano was reported by A. H. Delsemme (Acad. Roy. Sci. D'Outre-Mer, Bull. d. Seances, Bruxelles, new series, v. 6, p. 507-519, 1960) while this paper was in press. The identical result obtained for the Hawaiian and African volcanoes suggests that CuCl is a common fugitive constitutent of basaltic magmas.

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